Superconductivity in MgCNil₃

Sung-Ik Lee

Department of Physics, Sogang University, Seoul, Republic of Korea
Introduction of MgC\textsubscript{Ni} \textsubscript{3}

T. He et al. Nature 411, 54 (2001): \(T_c \sim 8\) K

\[ a \approx 3.812 \text{ Å} \text{ for } \text{MgC}_{0.96}\text{Ni}_3 \]

Simple cubic perovskite

\[ \text{High Ni-band near } E_F \Rightarrow \text{Expectation: ferromagnetism} \Rightarrow \text{In Reality, superconductivity} !!! \]
Introduction of MgCNi$_3$

Controversial issues in MgCNi$_3$

Issue of existence of spin fluctuation

Growth of MgCNi$_3$ single crystal

Transport, specific heat, tunneling properties of MgCNi$_3$ single crystal

Several evidences of conventional s-wave (electron-Phonon interaction) in MgCNi$_3$ single crystal

Conclusions
Controversial issues in MgCNI₃

- **Pairing symmetry**: 
  - *s-wave* \(^{13}\text{C NMR, specific heat, tunneling spectra)* or 
  - *non-s-wave* (tunneling spectra, penetration depth, critical current)

- **Electron-phonon coupling**: 
  - *moderate* (specific heat) or 
  - *strong* (specific heat, thermopower, tunneling spectra)

- **Charge carrier**: 
  - *hole* (theory) or 
  - *electron* (Hall, thermopower)
**Issue of pairing symmetry**

**s-wave:**

- $^{13}$C NMR (P. M. Singer…R. J. Cava, PRL 17 (2001))

  nuclear spin-lattice relaxation rate $1/ T_1$

  $\rightarrow$ coherence peak and exponential decrease

- Specific heat (J. Y. Lin…H. D. Yang, PRB 67 (2003))

  $C_{es}/\gamma nTc \approx 7.96\exp(-1.46Tc/T)$ and $\gamma(H) \propto H$

  cf.) non-s-wave: $\gamma(H) \propto \sqrt{H}$

- Tunneling (L. Shan…H. H. Wen, PRB 68 (2003))

  Andreev reflection spectra: well fitted by Blonder-Tinkham-Klapwijk (BTK) theory
non-s-wave:

- **Tunneling (Z. Q. Mao...R. J. Cava, PRB 67 (2003))**
  Zero-bias conductance peak (ZBCP)
  1) non-s-wave
  2) s-wave with multiband

- **λ (T) (R. Prozorov...R. J. Cava, PRB 68 (2003))**
  Tunnel-diode resonator
  \[ \Delta \lambda (T) \sim T^2 : d\text{-wave pairing with strong impurity scattering} \]
  cf.) clean d-wave: \[ \Delta \lambda (T)/\lambda (0) = T \ln 2/\Delta (0) \]
  s-wave: \[ \Delta \lambda (T)/\lambda (0) = \sqrt{\pi \Delta (0)/2T} \exp (-\Delta (0)/T) \]

- **Critical current (D. P. Young...P. W. Adams, PRB 70 (2004))**
  \[ J_c \propto [1-(T/T_c)^2]^\alpha \text{ where } \alpha = 2 : \text{unconventional} \]
  cf.) \( \alpha = 1.5 : \text{conventional} \)
Issue of electron–phonon coupling

electron-phonon coupling constant ($\lambda_{ph}$)

$\lambda_{ph} \ll 1$ : weak
$\lambda_{ph} < 1$ : moderate
$\lambda_{ph} > 1$ : strong

◈ Moderate

◆ specific heat

- T. He...R. J. Cava, Nature 411 (2001): $\lambda_{ph} = 0.77$
- J. Y. Lin ...H. D. Yang, PRB 67 (2003): $\lambda_{ph} = 0.83$
- Z. Q. Mao...R. J. Cava, PRB 67 (2003): $\lambda_{ph} = 0.94$
- L. Shan...H. H. Wen, PRB 68 (2003): $\lambda_{ph} = 0.83$

◈ Strong

◆ specific heat

- A. Wälte...L. Schultz, PRB 70 (2004): $\lambda_{ph} = 1.45$

◆ thermopower

- S. Y. Li...X. H. Chen, PRB 65, 064534 (2002): $\lambda_{ph} = 1.4 - 1.7$
Issue of charge carrier

- **Theory:** Majority hole band
- **Experiments:** electron

---

**Hall**

![Graph showing Hall effect](image)

**S. Y. Li et al., PRB 64 (2001)**

---

**Thermopower**

![Graph showing thermopower](image)

**S. Y. Li et al., PRB 65 (2002)**
Issue of existence of spin fluctuation

High and narrow DOS peak near $E_F$: a ferromagnetic instability!!

Expectation:
Ferromagnetism or Ferromagnetic spin fluctuation (SF)

For MgCNI$_3$ polycrystal
- Normal-state $\rho(T)$: No sign of SF
- $H_{c2}(T)$: Simple BCS

→ Evidences of SF in $\rho(T)$ and $H_{c2}(T)$ are still not yet clear.

FIG. 2. The total and atomic site projected local DOS of MgCNI$_3$. 

Mg: [Ne] 3s2
C: 1s2 2s2 p3
Ni: [Ar] 3d8 4s2
Electron scattering with

- **Ferromagnetic spin fluctuation:**
  - spin fluctuation
  - possibly $S = 1$

- **Magnon (quantized spin wave):**
  - Ferromagnetic spin wave
Evidences of ferromagnetic SF in NMR

- Spin susceptibility: Knight shift
- Spin fluctuations: Nuclear spin-lattice relaxation

MgC\text{Ni}_3

Saturation ~ 50 K

Saturation ~ 20 K

\begin{align*}
\chi_{\text{eff}}(T) &= \chi_{\text{sat}} - \chi_{\text{eff}}(0) \\
1/T_1(T) &= \left(\frac{\chi_{\text{eff}}(T)}{\chi_{\text{sat}}}ight)
\end{align*}

P. M. Singer \textit{et. al.} PRL 87 (2001)

Sr\textsubscript{2}RuO\textsubscript{4}

Down turn

T. Imai \textit{et. al.} PRL 81 (1998)
Theoretical $T_c$ suppressed by SF

No ferromagnetic SF ($\lambda_s = 0$) $\rightarrow$ $T_c > 20$ K of MgCNi$_3$

Modified McMillan equation

$$T_c = \frac{\Theta_D}{1.45} \exp \left( - \frac{1.04(1+\lambda)}{\lambda_p - \lambda_s - \mu^*(1+0.62\lambda)} \right), \quad \lambda = \lambda_p + \lambda_s$$

S. K. Bose, O. V. Dolgov, J. Kortus, O. Jepsen, and O. K. Andersen

$\lambda_p$ : Coupling constant of electron-phonon
$\lambda_s$ : Coupling constant of electron-paramagnon
$\mu^*$ : Coulomb potential

But, in reality, $T_c \sim 8$ K.

$\rightarrow$ SC and SF compete each other.

$\rightarrow$ SF suppresses $T_c$: $\lambda_s \neq 0$
### Comparison between MgCNi$_3$ and Sr$_2$RuO$_4$

<table>
<thead>
<tr>
<th>MgCNi$_3$</th>
<th>Sr$_2$RuO$_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ferromagnetically instable?</td>
<td>Ferromagnetically instable?</td>
</tr>
<tr>
<td>$T_c \sim 8$ K</td>
<td>$T_c \sim 1.5$ K</td>
</tr>
<tr>
<td>$H_{c2}(0) \approx 11$-$14$ T</td>
<td>$H_{c2}(0) \approx 1.5$ T</td>
</tr>
<tr>
<td>Simple cubic perovskite</td>
<td>Layered perovskite</td>
</tr>
<tr>
<td>pairing ?</td>
<td>$p$-wave (spin triplet pairing), mediates by ferromagnetic fluctuation</td>
</tr>
</tbody>
</table>
Comparison of RRR and $\rho_0$ with ferromagnetic Ni-compound

MgCNi$_3$ single crystal
- Superconductor
- $\rho_0 \approx 23 \ \mu\Omega cm$
- RRR $\approx 2.5$

Ni$_3$Al single crystal
- Ferromagnetic metal
- $\rho_0 \approx 1 \ \mu\Omega cm$
- RRR $\approx 2000$

→ Show strong scattering – Can we explain this from conventional BCS or Not
Claim of SF in MgCNI$_3$: upward curvature of $H_{c2}(T)$

$H_{c2}$: Upward Curvature near $T_c$  

cf. MgCNI$_3$ polycrystal
i) Inhomogeneity of the sample
   → Not proper ($\Delta T_c \sim 0.1$ K)

ii) Two-band nature
   → No evidential experiments

iii) Evidence of strong spin flip scattering
    As predicted by Y. N. Ovchinnikov and V. Z. Kresin,
    PRB 52 (1995)
Use of the polycrystalline MgCNi$_3$:

Strong scattering from defects and grain boundaries →

Screen intrinsic signal whether Magnon & SF exist

→ Need single crystals
Difficulties to synthesize MgCNi$_3$ single crystals

◈ Vast difference of melting temperatures: Mg (650 °C); Ni (1455 °C); C (3550 °C)

◈ Volatile Mg and less reactive graphite: Need excess Mg and C

⇒ We solved above problems while synthesizing in high-pressure.

Sung-Ik Lee, Advanced Materials 19(14), 1807 (2007)
Successful growth of the single crystalline MgCNi$_3$

Sung-Ik Lee, Advanced Materials 19(14), 1807 (2007)

Heat treatment at high pressure

single-crystalline MgCNi$_3$
EPMA (Electron Probe X-ray Micro Analyzer)

⇒ Mg : C : Ni = 1 : 1 : 2.8±0.1

Ni is slightly vacant

⇒ Well-defined simple cubic lattice structure with no notable secondary phase

⇒ a ~ 3.812 Å
Evidences of electron phonon mechanism from this study

- Normal-state $\rho(T)$ shows electron-phonon not magnon and SF scattering.

- A RRR and a much lower $\rho_0$ than poly → electron-phonon interaction

- $H_{c2}(T)$: linear in $T$ (simple BCS)

- Tunneling, Penetration depth → $S$-wave superconductivity

- Follows the theory of Electron-Phonon BCS Superconductivity not the Magnon & SF
\( \rho(T) \) at zero field in MgC\textsubscript{2}Ni\textsubscript{3} single crystal

- \( T_c \) (onset) = 6.7 K, 
  \( \Delta T_c \) (10-90\%) = 0.1 K
- \( \rho_0 \approx 23 \ \mu\Omega cm \)
- RRR = \( \rho(300 \text{ K})/\rho(8 \text{ K}) \approx 2.5 \)

For polycrystals,
- \( T_c \approx 6.4-8.4 \text{ K} \)
- \( \rho_0 \approx 40-1200 \ \mu\Omega cm \)
- RRR \approx 1.85-2.5
Field dependence of normal–state $\rho(T)$ of MgCNi$_3$ single crystal

- Conventional Behavior of resistivity
- Big Peak Effect
- $H_{c2}$ is linear in $T$ near $T_c$ $\rightarrow$ Conventional S-wave superconductor

Field dependence of normal–state $\rho(T)$ of MgC Ni$_3$ single crystal

$\rho(T)$ follows the Bloch-Gruneissen electron-phonon interaction

$H_{c2}$ is linear in $T$ near $T_c \rightarrow \rho(300K)/\rho(8K) = 2.5 > 1.85 \sim 2.5$ for poly

$\rho_0 \sim 23 \, \mu\Omega\text{cm} \ll \rho_0 \sim 40-1200 \, \mu\Omega\text{cm}$ for poly

MgC Ni$_3$ \text{good metal predicted by theory}
Superfluid density of MgCNi\textsubscript{3} single crystal from $H_{c1}$ and Tunneling diode $\rightarrow$ Conventional S-wave superconductor


\[ \rho_s(T) = \rho_s(0) \times e^{-T/T_c} \]

FIG. 3. (Color online) Normalized superfluid density deduced from $H_{c1}$ measurements (full symbols) and TDO measurements with $\lambda_0 = 230$ nm (open symbols) for samples A (squares) and C (circles). The solid lines are the fit for a superconducting gap $\Delta = 2k_BT_c$ with $T_c = 4.8, 5.6, 6.2,$ and $7$ K (see text). Inset: influence of the $\lambda_0$ value used to deduce $\rho_s$ from TDO measurements.
Values of energy gap $\Delta(0) \sim 1 - 1.2$ meV, with $2 \Delta /kT_c \sim 3.5 - 4.2$

Temperature dependences of two different point-contact spectra
Insets show $\Delta(T)$ of each contact and comparison to BCS-type (line)

$T_c \sim 6.8$ K
$\Delta(0) \sim 0.7$ meV

$2\Delta/kT_c \sim 2.5$ !!!

$T_c \sim 6.8$ K
$\Delta(0) \sim 1$ meV

$2\Delta/kT_c \sim 3.5$
Specific heat of MgCNi3

Total heat capacity plus addendum at zero field and background $C_n = g \cdot T + b \cdot T^3$

Electronic specific heat

$\Delta C(T)/T = C(T)/T - C_n(T)/T$

Entropy conservation: $\Delta C(T_c)/\gamma_n T_c \sim 1.7 - 1.8$

($\sim 1.43$ for BCS weak coupling limit)

From comparison with alpha model: $2 \Delta(0)/k_B T_c \sim 3.8$

($\sim 3.52$ for BCS weak coupling limit)

$\rightarrow$ a moderate-coupling superconductor

Single s-wave gap
Conclusions

- Successful growth of MgCNi\textsubscript{3} single crystal : $T_c \approx 7$ K, $\Delta T_c \approx 0.1$ K

- Evidences of Electron-Phonon interaction in MgCNi\textsubscript{3}
  1) $\rho(T)$ : follows from the electron-phonon scattering
  2) Lower RRR and larger $\rho_0$ than those of ferromagnetic metal
  3) $H_{c2}(T)$ for linear in $T$ near $T_c$ $\rightarrow$ conventional S-wave superconductivity
  4) Penetration depth $\rightarrow$ conventional S-wave superconductivity
  5) Tunneling and specific heat $\rightarrow$ moderate coupling conventional S-wave superconductivity
Abstract: Superconductivity in MgCNi$_3$

Sung-Ik Lee

Department of Physics, Sogang University, Seoul, Republic of Korea

In spite of the large number of ferromagnetic Ni atoms in its unit cell, MgCNi$_3$ shows superconductivity rather than Magnetism. According to band–structure calculations, MgCNi3 is near a ferromagnetic instability due to the high density of states at the Fermi level. However, whether or not the ferromagnetism in the formation of spin fluctuation remains is still unclear. The magnetism in the superconductivity can result in spin–triplet superconductivity or spin–affected conductivity. Moreover, the superconducting origin of this material has not yet been clearly identified, and the experimental results are still conflicting. For example, both s and non–s wave gap symmetry were observed by using various experimental tools. Also, the electron–phonon coupling constants in the conventional phonon–mediated pairing mechanism extracted from a number of specific heat measurements were different. Recently we successfully synthesized the MgCNi$_3$ single crystals and measured transport, penetration depth, tunneling and specific heat and clear the above issues. The origin of the superconductivity is from the electron–phonon interaction and gap symmetry is s–wave. Various issues will be discussed.