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Evidence of conventional superconductivity in single-crystalline $MgCNi_3$

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Abstract
Despite the strong expectation for a ferromagnetic ground state in $MgCNi_3$, the real ground state shows superconductivity without any evidence of magnetism. In the electrical transport measurements on single-crystalline $MgCNi_3$, we found that the ferromagnetic spin fluctuation either did not exist or was very suppressed and that $MgCNi_3$ could be well described by using the Bardeen–Cooper–Schrieffer theory. The evidence for this conclusion is as follows: (1) the normal-state resistivity could be explained by using the electron–phonon scattering model and (2) $H_{c2}(T)$ near T_c was linear.

1. Introduction

Ever since the discovery of $MgCNi_3$ [1], with its preference for superconductivity rather than the magnetism expected from the large number of magnetic Ni atoms in a unit cell, the origin of its superconductivity has been a mystery. For example, the pairing symmetry is still unclear. An s-wave pairing has been claimed on the basis of nuclear magnetic resonance (NMR) [2], tunneling spectroscopy [3], specific heat [1, 4–8], and lower critical field [9] measurements while a non-s-wave pairing has also been suggested on the basis of the behavior of other tunneling spectra [6], the penetration depth [10], and the critical current [11]. The charge carrier for this material is also controversial. Band calculations predict hole carriers [12–15] while Hall and thermopower measurements show electron carriers [16, 17]. To circumvent this issue, two-band superconductivity [8, 18] was proposed, but has not been well accepted because one band is enough to describe the specific heat and the upper critical field (H_{c2}).

One big issue with this compound is whether scattering from the spin fluctuation, in addition to the conventional electron–phonon pairing mechanism, exists [2, 7, 12–15, 19–21]. According to the calculated band structure for $MgCNi_3$, a high, narrow peak in the Ni 3d band just below the Fermi level could induce a ferromagnetic instability [12–15, 19–21], but so far, that instability has been reported only once, i.e., in the NMR data for polycrystalline $MgCNi_3$, where both the spin fluctuation effects and the spin-singlet superconductivity were observed [2]. In contrast to the NMR results, electrical transport observations [1, 8, 17, 22] for polycrystalline $MgCNi_3$ were explained by using a simple BCS theory without including the spin fluctuation. Since single crystals of $MgCNi_3$ are now available [23], this issue can be clarified by measuring the transport properties.

In this research, we measured the resistance as functions of the temperature and the applied magnetic field for single-crystalline $MgCNi_3$. We found that $MgCNi_3$ could be described by using the conventional electron–phonon pairing mechanism, excluding the spin fluctuation, which is in contrast to the theoretical prediction. The evidence for this is as

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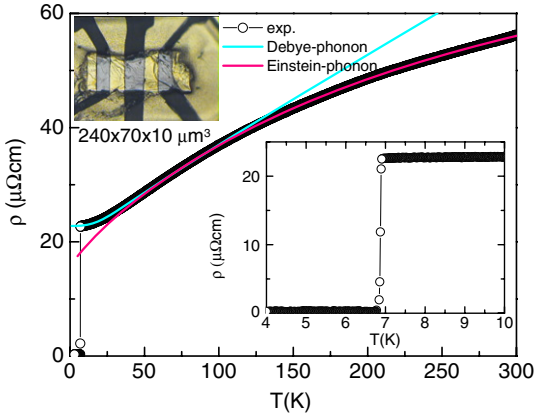

Figure 1. Temperature dependence of the resistivity at zero field for the $MgCNi_3$ single crystal. Upper inset: an image of the sample with four-probe contact leads. Lower inset: a magnified view of the $\rho(T)$ near T_c. The open circles are the experimental data, and the blue and the pink solid lines are the theoretical fitting curves obtained by using the Debye-phonon model and the Einstein-phonon model, respectively. The details are in the text.

(This figure is in colour only in the electronic version)

follows: (1) the normal-state resistivity ($\rho(T)$) could be explained by using only electron–phonon scattering and (2) $H_{c2}(T)$ near T_c showed a linear behavior, as predicted in the BCS theory.

2. Experimental details

For several years after the discovery of superconductivity in $MgCNi_3$, the growth of single crystals was a formidable task. Compared to the range of synthesis conditions for MgB_2 single crystals, which were synthesized by several groups at high pressure just after the discovery of its superconductivity, that for $MgCNi_3$ turned out to be very narrow. Only very recently, inside a high-pressure furnace, have single crystals of several hundred microns in size been grown [23]. By using an electron probe x-ray microanalyzer, we found the deficiency of carbon with respect to stoichiometry for our single crystals to be negligible. The x-ray diffraction pattern and the transmission electron microscopy image showed that the $MgCNi_3$ single crystals had a simple cubic crystal structure. For the transport measurements, we selected clean, flat single crystals with sizes of a few hundred micrometers and fabricated four metallic leads by using a photolithographic technique. An image of the sample with four leads is shown in the upper inset of figure 1. The temperature (T) and the magnetic field (H) dependences of the resistivity (ρ) were measured by using a standard DC four-probe method. Crystals from the same batch showed the same transport properties.

3. Results and discussion

Figure 1 shows the $\rho(T)$ of the $MgCNi_3$ single crystal without a magnetic field applied. The T_c onset is about 6.9 K, as shown in the lower inset of figure 1. The transition width of $\Delta T_c \sim 0.05$ K, which was determined by using a criterion of 10–90% of the normal-state ρ, is very sharp. The normal-state $\rho(T)$ curve of the single crystal has an upward curvature at low temperatures and a downward curvature at higher temperatures, which is similar to the situation reported for polycrystals. The residual resistivity ratio RRR $= \rho(300\ \mathrm{K})/\rho(8\ \mathrm{K})$ is about 2.5, which is larger than that of polycrystals (1.85–2.5) [1, 8, 17, 22]. The residual resistivity of the crystal, $\rho_0 \simeq 23\ \mu\Omega$ cm, is smaller than that of polycrystals (40–1200 $\mu\Omega$ cm) [1, 8, 17, 22]. This indicates that impurity scattering does not dominate the intrinsic signal in $MgCNi_3$ single crystals. However, the relatively low RRR and the relatively high ρ_0 of the single crystal compared to those of good metals indicate that $MgCNi_3$ has poor metal properties at the normal state, as predicted by theory [19].

The resistive behavior of the normal state could be well described by using the Bloch–Grüneisen model in which electrons are scattered by phonons with two different modes. In the Debye-type phonon model (an acoustic mode) [24],

$$\rho(T) = \rho_0 + \rho_{\mathrm{ph}}(T),$$
$$\rho_{\mathrm{ph}}(T) = \rho_1 \left(\frac{T}{\Theta_D}\right)^3 \int_0^{\Theta_D/T} \frac{z^3\,\mathrm{d}z}{(1-\mathrm{e}^{-z})(\mathrm{e}^z-1)}, \quad (1)$$

where ρ_0 is the temperature-independent residual resistivity, $\rho_{\mathrm{ph}}(T)$ is the phonon scattering contribution, ρ_1 is a proportionality constant, and Θ_D is the Debye temperature. In the Einstein-type phonon model (an optical mode) [25, 26],

$$\rho(T)^{-1} = \rho_p^{-1} + (\rho_0 + \rho_{\mathrm{ph}}(T))^{-1},$$
$$\rho_{\mathrm{ph}} = \rho_l \coth(\Theta_E/2T)[1 + (2/3)\sinh^2(\Theta_E/2T)]^{-1}, \quad (2)$$

where ρ_p is the parallel part of the resistivity and Θ_E is the Einstein temperature. The experimental data were well fitted by (i) the Debye-phonon model at lower temperatures (blue line in figure 1) and by (ii) the Einstein-phonon model at higher temperatures (pink line in figure 1). The parameters obtained are $\Theta_D \simeq 132$ K, $\rho_0 \simeq 23\ \mu\Omega$ cm, and $\rho_l \simeq 40\ \mu\Omega$ cm in case (i) and $\Theta_E \simeq 223$ K, $\rho_0 \simeq 39\ \mu\Omega$ cm, $\rho_l \simeq 31\ \mu\Omega$ cm, and $\rho_p \simeq 105\ \mu\Omega$ cm in case (ii).

According to inelastic neutron scattering measurements by Heid *et al* [27] and calculations of lattice dynamics by Wälte *et al* [8] and Ignatov *et al* [28], the phonon density of states of $MgCNi_3$ is dominant in Ni modes, and Ni optical branches vibrate around phonon temperatures of 185 K [27] and 250 K [8, 28]. Here, we estimated the value of 185 K from the strong main peak of the Ni phonon density of states around 16 meV. Our Einstein temperature of 223 K is in the range of those values. From this result, we found that the electrons of $MgCNi_3$ in the normal state are scattered by Ni-based optical phonons.

The Einstein-type phonon model, which has a lower slope of resistivity than the Debye-phonon model, gives the downward behavior just after the upward curvature in $MgCNi_3$. However, the Einstein-phonon model gives a nearly linear behavior of the resistivity at high temperatures. Thus, the parallel part of the resistivity in equation (2) causes the slope to decrease more at higher temperatures near 300 K in $MgCNi_3$. Several models including the parallel resistance have been

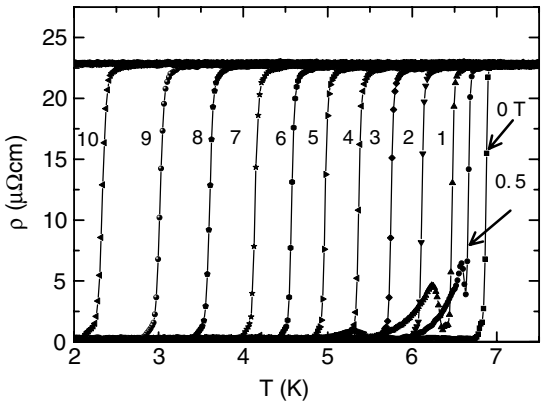

Figure 2. Temperature dependence of the resistivity for the $MgCNi_3$ single crystal at different fields.

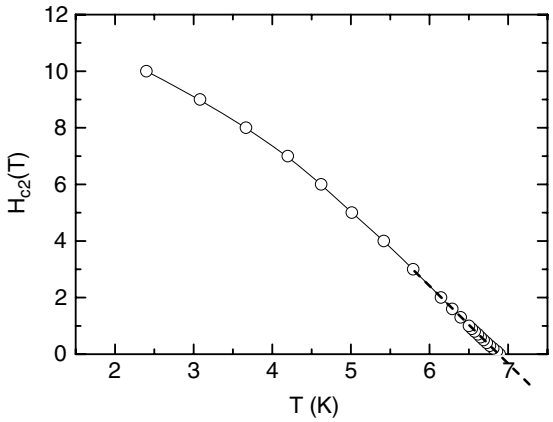

Figure 3. Upper critical field determined at 90% of the normal-state ρ, as shown in figure 2.

proposed for describing the decreasing slope [29, 30]. The negative curvature at higher temperatures is reminiscent of resistivity saturation, which is well observed in many metallic systems. The saturation occurs when the mean free path of the charge carriers becomes comparable to the interatomic distance [29, 30]. According to $\rho = \frac{3\pi^2\hbar}{e^2k_F^2l}$, assuming a three-dimensional system having a spherical Fermi surface [30], the mean free path l for $\rho \approx 56\ \mu\Omega$ cm at 300 K in $MgCNi_3$ is about 51 Å, which is much larger than the interatomic separations, indicating that the resistivity at 300 K is still approaching saturation. Here, we used the conversion $\hbar a_0/e^2 = 0.022$ mΩ cm, the Bohr radius $a_0 = 0.529$ Å, and the Fermi wavevector $k_F = \sqrt{2\pi}/a$, where a is the lattice parameter.

The values of Θ obtained from our experiment are lower than the $\Theta_D \geqslant 284$ K obtained from specific heat measurements on polycrystals [4–8]. However, using the value of $\Theta_E \simeq 223$ K and assuming an electron–phonon coupling strength of $\lambda \simeq 0.67$ and a Coulomb pseudopotential of $\mu^* = 0.1$ in the McMillan formula refined by Allen and Dynes [31],

$$T_c = \frac{\Theta}{1.45}\exp\left[\frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right], \tag{3}$$

in which magnetic scattering is not included, we could estimate the T_c of $MgCNi_3$ to be about 6.9 K, which is in good agreement with the T_c for our $MgCNi_3$ single crystals. Here, $\lambda \simeq 0.67$ is in the range of $\lambda \simeq 0.6$–0.8, which was obtained from recent specific heat measurements [32] using our $MgCNi_3$ single crystals, and μ^* is usually 0.1–0.15 in the absence of spin fluctuations, as presented in [28].

From the above-mentioned results, there is a crossover at around 70 K that is induced by a change in the phonon modes. The origin of the unusual shape of $\rho(T)$ for $MgCNi_3$ is the Debye-phonon contribution for lower T and the Einstein-phonon contribution for higher T. In particular, we did not observe the spin fluctuation effect, which is of particular interest for $MgCNi_3$. This implies that $MgCNi_3$ is a conventional, phonon-mediated superconductor with intermediate coupling strength.

Figure 2 shows the temperature dependence of the resistivity of $MgCNi_3$ at different magnetic fields. The peak effect, which is presented only for clean and weakly pinned single crystals such as $NbSe_2$ [33–36], BSCCO [37, 38], and MgB_2 [39–42], appears. As figure 2 shows, the superconducting transitions under magnetic fields are very sharp; thus, the H_{c2} value does not depend very much on the criterion used to determine it. The $H_{c2}(T)$, which was determined at 90% of the normal-state ρ, is shown in figure 3. The $H_{c2}(T)$ near T_c follows the linear dotted line. The fact that the linearity of $H_{c2}(T)$ is predicted by the BCS theory without considering magnetic scattering strongly supports the notion that magnetic scattering does not occur in $MgCNi_3$.

Within the BCS theory, $H_{c2}(T = 0)$ can be estimated by using the Werthamer–Helfand–Hohenberg (WHH) formula [43]:

$$H_{c2}^{WHH}(0) = -0.693T_c\left(\frac{dH_{c2}}{dT}\right)_{T_c}, \tag{4}$$

which leads to $H_{c2}^{WHH}(0) \approx 12.8$ T, where the slope $(dH_{c2}/dT)_{T_c}$ is around -2.7 T K^{-1}. Meanwhile, the Pauli-limiting field [44],

$$H^{Pauli} = 1.24k_BT_c/\mu_B, \tag{5}$$

is about 12.7 T. H_{c2}^{WHH} and H^{Pauli} are almost the same. The superconducting coherence length $\xi(0)$ is estimated to be approximately 51 Å by using $H_{c2}(0) = \Phi_0/2\pi\xi^2(0)$.

4. Conclusion

Although a ferromagnetic instability in the Ni-based material $MgCNi_3$ has been predicted, up to now the existence of a spin fluctuation or ferromagnetism has been controversial. Now, on the basis of several pieces of evidence from electron scattering with phonons in $MgCNi_3$ single crystals, we conclude that the fluctuating spins either do not contribute to the transport properties or are very suppressed. The linear temperature dependence of H_{c2} near T_c also strongly supports this idea. Finally, our transport measurements for $MgCNi_3$ single crystals support the superconducting origin for $MgCNi_3$ being a simple electron–phonon coupling.

Acknowledgments

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