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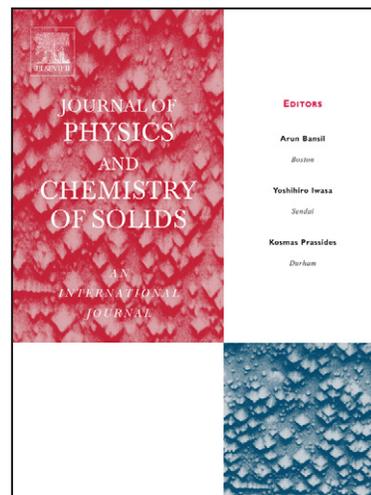
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Superconducting energy gap in MgCNi₃ single crystals

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Abstract

Even though MgCNi₃ contains large molar volume of magnetic Ni atoms it clearly shows superconductivity. The origin of superconductivity in this material has not yet been clarified. There is a lot of discrepancy in experimental results and physical interpretation where even unconventional pairing or a two-band model have been proposed for the system. All that is based on results obtained on polycrystalline samples. Here we present the first point-contact spectroscopy and ac-calorimetry measurements on the MgCNi₃ single crystals of very good quality. Measurements have been performed in the temperature range from 1.5 to 10 K and in magnetic fields up to 8 T. The temperature dependence of the energy gap of the system is presented and compared to the BCS model.

1. Introduction

Despite the fact that MgCNi_3 contains also magnetic Ni ions it shows superconductivity around 7 K [1]. The origin of superconductivity remains unclear. Mao *et al.* [2] have discussed unconventional pairing based on their point-contact data showing the zero-bias peak. Another point-contact experiments on MgCNi_3 [3] have also found the zero-bias anomaly in the tunneling conductance data but explained it not as a consequence of unconventional pairing mechanism but due to an extrinsic origin, namely due to a presence of weak links near the point contact. With this assumption the BCS *s*-wave gap has been obtained from fitting of the data. The measurements of the penetration depth [4] have indicated non-*s*-wave superconductivity. The heat capacity and the upper critical field data [5] have been described by a single-band/single-gap *s*-wave model with strongly coupled holes. In order to explain the Hall conductivity and thermopower data published in the same paper the two-band/two gap model of phonon mediated *s*-wave superconductivity partly suppressed by an electron-paramagnon interaction has been proposed. To resolve this controversial situation measurements on single crystals of good quality are highly desirable.

Recently, the MgCNi_3 crystals of high quality have been grown [6]. Here we present the first point-contact spectroscopy and ac-microcalorimetry data on these crystals with aim to study the superconducting energy gap in the system. Although certain scattering of the gap value is found in our point-contact spectra, which is probably due to the surface inhomogeneity, these results together

with the analysis of our heat-capacity measurements suggest single gap *s*-wave superconductivity with the medium coupling in the system.

2. Experimental

Difficulties in preparation of MgCNi_3 single crystals have been overcome using a high-pressure closed system as described elsewhere [6]. X-ray analysis proved that carbon deficiencies from stoichiometry are negligible. Contrary to the polycrystalline MgCNi_3 samples, which have usually local carbon deficiency, in the single crystals the Ni sites were partly deficient. This leads to a critical temperature $T_c \sim 6.7$ K (obtained from transport measurements) which is lower than the highest T_c of 7.3 K of polycrystalline samples. The samples used for our measurements have typical dimensions of few hundred microns.

The point-contact spectroscopy measurements were performed on several samples from the same batch. A standard lock-in technique was used to measure the differential resistance as a function of applied voltage on the point contacts. The point contacts were prepared *in situ* by mechanically pressing platinum tips on the surface of the crystals. The approaching system allows for both lateral and vertical movements of the tip enabling a formation of the junction as well as the probing of different places on the sample surface. The point-contact differential conductances (calculated from differential resistance) have been normalized to the conductance background found at the voltages above the gap. The normalized curves were fitted to the standard point-contact model accounting for the Andreev reflection as well as the tunneling contributions to the point-contact current. This

allows obtaining information about the superconducting energy gap(s) Δ , the spectral broadening Γ and the barrier transparency z [7].

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. The ac microcalorimeter [8] which uses a diode as the heater has been used. 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 very sensitive thermocouple.

The point-contact and ac heat capacity measurements of MgCNi_3 single crystals were performed in the temperature range between 2 and 10 K and in magnetic fields up to 8 T.

3. Results and Discussion

Tens of the point-contact spectra and few sets of their temperature dependences have been recorded on several crystals from the same batch. The point contacts revealed intermediate barrier strength with both Andreev reflection and tunneling contributions. The spectra have shown a well defined pair of peaks indicating the size of the superconducting energy gap. By positioning the tip at different places on the surface of the samples a certain scattering of the gap peaks was observed in line with the previous observations [3]. The scattering could be due to local inhomogeneities, e.g. local phases with slightly different T_c 's, so it was important to know the local transition temperature at the junction. For this, the temperature dependence of the point-contact spectrum was measured. Then, the point-contact spectra were fitted to the point-contact s-wave formula accounting also for

the temperature smearing. The gap value as a function of temperature was resulting from the fit. The superconducting energy gap extrapolated to zero temperature $\Delta(0)$ was found to be scattered between 1.0 and 1.2 meV for different measurements although the local T_c equal to 6.6 ± 0.1 K was quite homogeneous. Figure 1 shows such an example with the inset showing the temperature dependence of the gap together with the BCS prediction. The resulting coupling ratio of $2\Delta(0)/k_B T_c$ in the range between to 3.5 and 4.2 was found. In some cases a significantly smaller size of the gap $\Delta(0)$, about 0.7 meV was also detected. Beside the small size of this gap, its temperature dependence revealed a significant deviation from the BCS shape. Namely, although the local T_c has been again in the range of 6.6 ± 0.1 K, the measured gap undershooting the BCS prediction in the temperature range between 3 K and T_c . Such behavior is indicative for a proximity effect on the surface with somewhat degraded superconductivity. There, the degraded surface layer has a lower T_c^* and smaller $\Delta^*(0)$ which has a tendency to close at T_c^* (undershooting $\Delta(T)$ with the bulk T_c), but due to an increase of the coherence length near the bulk T_c the superconductivity is kept until T_c where Δ^* is finally closed.

Another possible interpretation of observing smaller and larger gaps would be a two gap scenario. We remark that the two-gap behavior can be effectively unveiled when magnetic field is applied on point contacts [7]. Details of the gap measurements on MgCNi_3 at different crystallographic directions as a function of temperature and magnetic field will be published elsewhere [10] but these results point again to the single gap scenario [10].

Thus, from the point-contact spectroscopy we can conclude that MgCNi_3 behaves like an *s*-wave single-gap superconductor with weak up to moderate coupling strength.

Since the point-contact spectroscopy is a surface sensitive technique it is important to compare its results with the bulk thermodynamic measurements such as the heat capacity. The samples have shown a well-defined superconducting transition in the heat capacity. The anomaly represents around 40% of the total signal including addenda. It is sharper ($\Delta T_c \sim 0.2$ K) than the specific heat anomaly observed on the polycrystalline samples [6], indicating the quality and homogeneity of our samples. To clarify the superconductivity in MgCNi_3 , it is of interest to derive the electronic heat capacity by subtracting the normal state heat capacity C_n i.e. $\Delta C(T)/T = C(T)/T - C_n(T)/T$. A magnetic field of 8 T was enough to suppress the superconductivity down to 3 K in our case. At low temperature region the high field heat capacity $C(8\text{T})$ with subtracted addenda has followed well the usual formula $C(8\text{T})/T = C_n/T = A + B.T^2$. This dependence was extrapolated to the lowest temperatures. The value of $\Delta C(T_c)/\gamma_n T_c$ can be used to measure the strength of the electron coupling. With the heat capacity data measured down to 2.4 K γ_n could not be determined directly. To estimate γ_n a plot of $\Delta C(T)/T$ dependence on temperature down to 0 K was made following the entropy conservation law, demanding that the areas between the superconducting and normal part below and above the crossing point must have equal surfaces. This procedure will be verified by the experiments in He-3 cryostat which are in progress.

The resulting $\Delta C(T)/T$ at $H=0$ is shown in Fig. 2. From an entropy conserving construction (see the dotted line), the transition temperature T_c was estimated to be 5.86 K. We remark that this T_c is lower than that deduced from the transport and point-contact measurements on the samples from the same batch.

The obtained ratio of $\Delta C(T_c)/\gamma_n T_c$ estimated from our data is equal to 1.71. This value is larger than the BCS weak coupling limit (1.43) indicating stronger coupling. Another possibility to estimate

the coupling strength of MgCNi_3 is to compare the measured dependence of $\Delta C(T)$ to the so called alpha model [9] based on the BCS theory. In this model the only adjustable parameter is the gap ratio $2\Delta(0)/k_B T_c$. In our case the comparison yields a ratio $2\Delta(0)/k_B T_c \sim 3.8$, which is higher than the BCS weak coupling limit 3.52 in agreement with the large value of $\Delta C(T_c)/\gamma_n T_c$. Thus, the heat capacity measurements values also suggest that the MgCNi_3 may be a superconductor with a moderate coupling strength. The measurements at lower temperatures capable to resolve the symmetry of the superconducting gap are in progress.

3. Conclusions

We have performed the point-contact spectroscopy and highly sensitive ac-microcalorimetry studies on MgCNi_3 single crystals. Both methods point to the conventional s-wave single-gap superconductivity with the moderate coupling in the system.

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REFERENCES

- [1] T. He et al., Nature **411**, 54 (2001).
- [2] Z.Q. Mao et al., Phys. Rev. B **67**, 094502 (2003).
- [3] L. Shan et al., Phys. Rev. B **68**, 144510 (2003).
- [4] R. Prozorov et al., Phys. Rev. B **68**, 180502 (2003).
- [5] A. Wälte et al., Phys. Rev. B **70**, 174503 (2004).
- [6] H-S. Lee et al., Adv. Mater. **19**, 1807 (2007).
- [7] P. Szabó et al., Phys. Rev. Lett. **87**, 137005 (2001).
- [8] P.F. Sullivan and G. Seidel, Phys. Rev. **173**, 679 (1968).
- [9] H. Padamsee et al., J. Low Temp. Phys. **12**, 387 (1973).
- [10] Z. Pribulová et al., to be published.

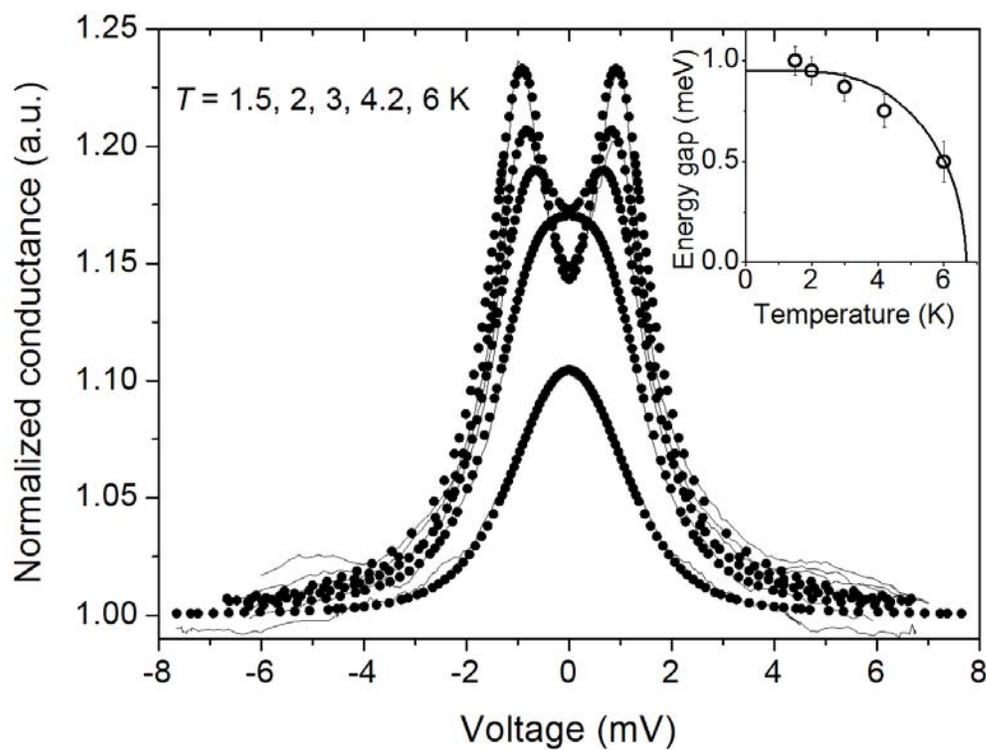


Fig. 1 Pt- MgCNi_3 point-contact spectra (solid lines) measured at indicated temperatures and fit to the single gap s -wave point-contact formula (circles). Inset: Temperature dependence of the fitted energy gap with $\Delta(0) = 1$ meV and the BCS prediction (line).

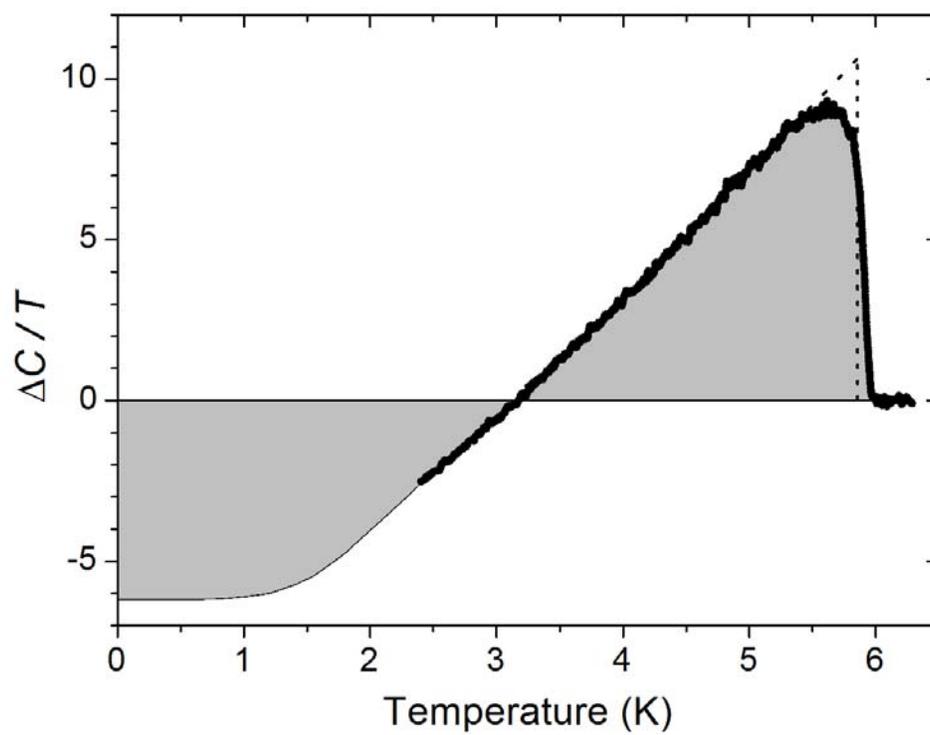


Fig. 2 Electronic heat capacity in relative units. Dotted lines indicates the transition temperature as determined by conservation of entropy.