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Muon spin rotation study of $MgCNi_3$

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Abstract

We have performed transverse field μSR measurements of the non-oxide perovskite compound $MgCNi_3$ down to 20 mK to determine the magnetic field penetration depth, finding $\lambda(0) = 2315 \pm 10$ Å. We have also performed zero-field measurements and can exclude any spontaneous internal fields greater than 0.02 G. Thus, we conclude that the superconducting state does not possess broken time reversal symmetry.

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1. Introduction

Since the discovery of superconductivity in $MgCNi_3$ by He et al. in 2001 [1], there has been a flurry of activity around this compound [2–6,8–14]. Originally investigated as a three-dimensional (3-d) analogue of the layered $LnNi_2B_2C$ family of superconductors, it quickly became apparent that there were several other intriguing properties. Its structure resembles both the cuprates and several conventional superconductors. It has several bands near the Fermi surface, giving it a complicated electronic structure. It has a variable carbon content which strongly affects the superconducting state.

Most provocative though is the possibility of an unconventional superconducting mechanism involving ferromagnetic spin fluctuations, as first discussed in the original paper when the authors attracted attention to the high nickel content of the material [1]. Band structure calculations note the presence of a large peak of mainly Ni 3-d character in the density of states, located just below the Fermi surface [2,3]; photoemission and absorption experiments have confirmed such a peak [4]. This has led to predictions of a quantum phase transition to a ferromagnetic ground state with slight hole doping [2,3]. Indeed, it has been observed that superconductivity disappears with a mere 1% doping of Co on the Ni site [5] or 12% carbon vacancies [6], but no ferromagnetism has been observed at any composition to date. Nonetheless, the possibility remains that magnetism plays a central role in determining the properties of the superconducting state.

Historically, strong magnetic interactions in superconducting systems have often manifested themselves in anisotropic pairing, as seen in many of the heavy fermion superconductors [7]. As such, several experiments have been performed to search for line nodes in superconducting gap of $MgCNi_3$. Unfortunately, the results of these experiments have been unclear and often contradictory.

Specific heat measurements seem to indicate a fully developed s-wave gap, but contradict each other when determining coupling strength and the role of spin fluctuations [8,9]. A zero-bias conductance peak seen in point contact tunneling was long taken as evidence for line nodes in the gap [10], but has been possibly explained by conventional Andreev reflection [11]. A T^2 temperature dependence of the magnetic penetration depth was

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observed by microwave resonance [12], which is strong evidence for a gap with nodes. However, a Hebel–Schlichter peak was seen in NMR [13], which is equally strong evidence for an s-wave gap. No concensus has yet been reached.

Voelker and Sigrist [14] have put forth a theory that attempts to reconcile these seemingly disparate experiments, wherein a multi-band s-wave order parameter creates unconventional behaviour through inter-band scattering. However, the theory firmly predicts the existence of spontaneous time reversal symmetry breaking (TRSB) in the superconducting state. Such symmetry breaking should create spontaneous internal magnetic fields, which could be detected.

Muon spin rotation is in a good position to clarify the situation with $MgCNi_3$. Through the use of transverse field μSR (TF-μSR) in the vortex state of the superconductor, one can obtain a measure of the local penetration depth which does not depend on surface effects [15]. The temperature-dependence of this penetration depth is a sensitive measure of the symmetry of the order parameter [16]. Also, with zero field μSR (ZF-μSR), one can look for the emergence of spontaneous local magnetic fields, a sign of TRSB. Such experiments have successfully identified TRSB states in exotic superconductors in the past [17,18].

In this paper, we report the results of TF-μSR and ZF-μSR experiments on a ceramic sample of $MgCNi_3$ at TRIUMF in Vancouver, Canada. We will present the data and discuss their implications on the superconducting state in this material.

2. Sample characterisation

A ceramic sample of nominally stoichiometric $MgCNi_3$ was synthesised at Princeton University. X-ray diffraction experiments were performed on a Bruker D8 powder diffractometer at McMaster University. The resulting diffraction pattern is well described by the perovskite $MgCNi_3$ structure and reveals that the sample is 98.7% pure by weight. The only observed impurity is MgO, presumably from excess Mg added during the initial reaction [1]. MgO is nonmagnetic, and should have no effect on a μSR experiment.

Magnetisation was measured in a Quantum Design MPMS SQUID magnetometer on two separate pieces of the ceramic to test sample homogeneity. Following the convention laid out in Ref. [6], we define T_c as the temperature at which the steepest part of the magnetisation versus temperature curve intersects the x-axis. The resultant critical temperatures were $T_c = 7.05 \pm 0.01$ and $7.07 \pm 0.02\,K$, respectively. The two values are well within each other's uncertainty range, confirming that the bulk properties are uniform across the sample. A critical temperature of 7.06 K corresponds to a carbon occupation of $x \sim 0.976$ [6].

3. TF-μSR

TF-μSR measurements were performed on both the Lampf spectrometer on the M20 beamline and the dilution refrigerator on the M15 beamline of TRIUMF in Vancouver, BC.

Measurements were performed in a field of 2 kG, which is well into the vortex state of the superconductor but much less than its upper critical field. Preliminary inspection of the FFT of the TF-μSR spectra show an asymmetric lineshape, confirming that we are indeed probing a well-formed vortex lattice [15]. However, imperfections in the vortex lattice were prevalent enough to broaden many of the features of the Abrikosov field distribution, and attempts to fit the spectra to an appropriate vortex lineshape have failed. Instead, spectral envelopes were approximated by a simple Gaussian ($e^{-\sigma^2 t^2/2}$), whose width still reflects the superfluid density. Fig. 1 displays the envelope relaxation rate as a function of temperature.

The dashed line in Fig. 1 shows a fit to the weak coupled BCS prediction. The curve seems to qualitatively capture the behaviour of the relaxation but is statistically poor, being systematically high or low at almost every temperature. The fit also predicts a transition temperature of $T_c \approx 7.38 \pm 0.01\,K$, inconsistent with the value of 7.06 K obtained from magnetisation measurements. This suggests that if the superconductor is s-wave, it is not weakly coupled. Also shown is the superfluid density for $YBa_2Cu_3O_{6.99}$ [16] (solid curve), a well-known d-wave superconductor. Comparison of this curve with our data allows us to safely eliminate any strong linear decrease in n_s as seen in clean nodal superconductors.

Prozorov et al. [12] have recently observed a T^2 dependence in their measurements of penetration depth and argued that this is indicative of a superconductor with line nodes and prominent impurity scattering. Fig. 2 shows a comparison of our data with that of Prozorov et al.,

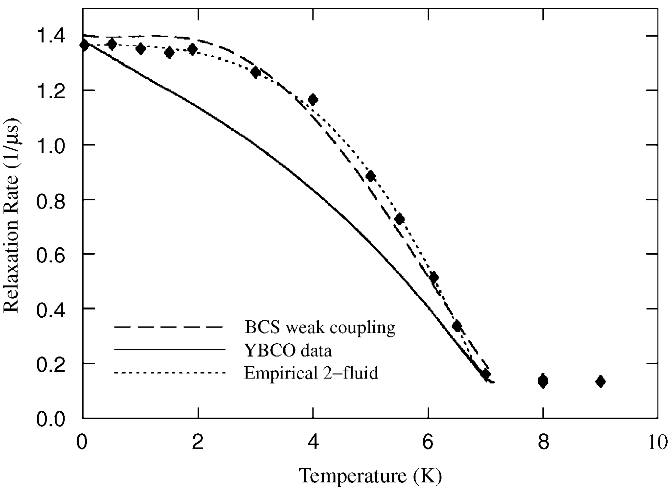

Fig. 1. Relaxation rates obtained from Gaussian fits to TF-μSR spectra. Curves are described in the text.

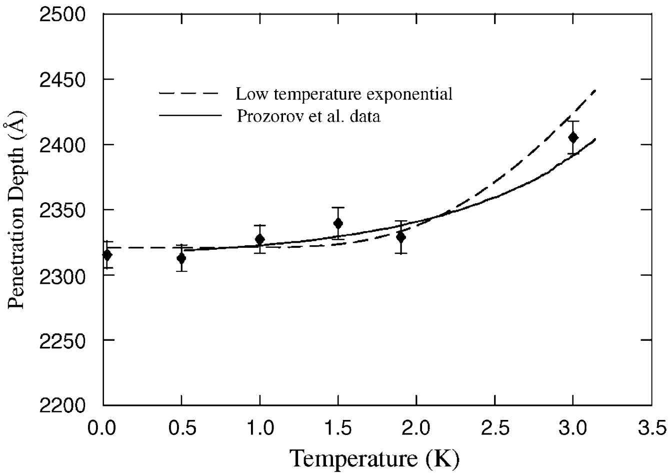

Fig. 2. Penetration depths as a function of temperature. Curves are described in the text.

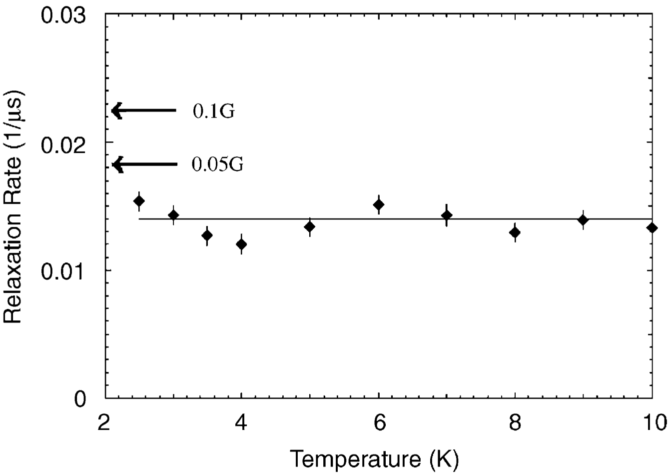

Fig. 3. The relaxation rate versus temperature for the ZF-μSR experiments. Arrows indicate where one would expect the curve to rise to for the TRSB fields indicated.

properly normalised. Also plotted is a fit to the low-temperature expression expected for a fully developed s-wave gap, with fit parameter $\Delta(0)/T_c = 1.7 \pm 0.3$. This value for the gap ratio is within that expected for weak coupled BCS theory and much higher than that extracted from fits to the Prozorov data. As one can see, our data is consistent with both a BCS gap and Prozorov's data, and cannot be used to distinguish between the two.

In an effort to characterise our relaxation curve, we have also fit to an empirical "two fluid model" of the form $n_s = n_0(1 - (T/T_c)^m)$, as represented by the dotted line in the Fig. 1. The curve describes the data well over the entire temperature range with fit parameters $m = 3.03 \pm 0.06$ and $T_c = 7.08 \pm 0.02$ K. Note that the transition temperature is now consistent with our magnetisation measurements. As the fit range is restricted to only lowest temperatures ($T \leqslant 2$K), a wider range of exponents are also consistent with the data, including that obtained by Prozorov.

Using the relation $\lambda = 2700 \times \sigma^{-1/2}$ [19], we obtain $\lambda(0) \approx 2315 \pm 10$ Å for the absolute value of magnetic penetration depth. This value is approximately 7% lower than previous values obtained from critical field [10] and magnetisation measurements [12], but within error bars of the latter. One should note, however, that measured values of λ can vary up to 15% depending on one's model for field distribution and relaxation function.

4. ZF-μSR

We also performed ZF-μSR measurements on the Lampf spectrometer to search for internal fields characteristic of a TRSB state. As sensitivity to internal magnetic fields is ultimately limited by the observed background field, we first went to great lengths to minimise the field at the sample site due to external sources. This was done by using trim coils to minimise the muonium precession-frequency in a single crystal of high-purity silicon, as per the technique of Morris and Heffner [20]. By such a method, we managed to zero the local magnetic field in each direction to less than 40 mG.

We then measured zero field spectra for $MgCNi_3$ as a function of temperature. Spectra were well described by a single component exponential decay. This represents inhomogeneous relaxation due to diffuse moments of ^{25}Mg, which composes 10% of the natural abundance of Mg. The temperature dependence of the relaxation rate is shown in Fig. 3.

If there were TRSB in the superconducting state as predicted by Voelker and Sigrist, one would expect to see a rise in the relaxation rate below the critical temperature, corresponding to an increase in the average internal field size. The arrows in Fig. 3 show the point to which we would expect the curve to rise for average spontaneous fields of amplitude 0.05 and 0.1 G, respectively. Instead, we see no increase in relaxation at T_c or any other temperature down to 2 K.

The scatter in the data points reflects our experimental uncertainty, and indicates the maximum size of TRSB fields which could exist and escape our detection. Taking our uncertainty to be one standard deviation from a flat line, we arrive at a maximum internal field value of ~0.02 G. This is comparable to the uncertainty in fields from external sources, and is more than an order of magnitude less than to the value of 0.5 G which was observed in the TRSB state of Sr_2RuO_4 [17]. Thus, we have effectively ruled out TRSB in this material.

5. Conclusions

In conclusion, we have performed TF-μSR and ZF-μSR experiments as a function of temperature on a polycrystalline sample of $MgCNi_3$. Transverse field relaxation measurements give $\lambda(0) \approx 2315 \pm 10$ Å. Empirical fits reveal a T^3 decrease in superfluid density over the entire temperature range. ZF-μSR measurements put an upper limit of ~0.02 G on any possible TRSB fields in the superconducting state.

Acknowledgements

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