Temperature Dependence of the Magnetic Penetration Depth in the Vortex State of the Pyrochlore Superconductor, $Cd_2Re_2O_7$

M. D. Lumsden,[1] S. R. Dunsiger,[2] J. E. Sonier,[3] R. I. Miller,[4] R. F. Kiefl,[4,5,9] R. Jin,[1] J. He,[6] D. Mandrus,[1,6] S. T. Bramwell,[7] and J. S. Gardner[8]

[1]*Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831*
[2]*Los Alamos National Laboratory, MST-10, MS K764 Los Alamos, New Mexico 87545*
[3]*Department of Physics, Simon Fraser University, British Columbia, Canada V5A 1S6*
[4]*Department of Physics and Astronomy, University of British Columbia, Canada V6T 1Z1*
[5]*TRIUMF, Vancouver, British Columbia, Canada V6T 2A3*
[6]*Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996*
[7]*Department of Chemistry, University College London, London WC1H OAJ, United Kingdom*
[8]*National Research Council, NPMR, Chalk River Laboratories, Chalk River, Ontario, Canada K0J 1J0*
[9]*Canadian Institute for Advanced Research, 180 Dundas Street West, Toronto, Ontario, Canada M5G 1Z8*

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We report transverse-field and zero-field muon spin rotation and relaxation studies of the superconducting rhenium oxide pyrochlore, $Cd_2Re_2O_7$. Transverse-field measurements ($H = 0.007$ T) show line broadening below T_c, which is characteristic of a vortex state, demonstrating conclusively the type-II nature of this superconductor. The penetration depth is seen to level off below about 400 mK ($T/T_c \sim 0.4$), with a rather large value of $\lambda(T = 0) \sim 7500$ Å. The temperature independent behavior below $\sim$400 mK is consistent with a nodeless superconducting energy gap. Zero-field measurements indicate no static magnetic fields developing below the transition temperature.

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The pyrochlore transition metal oxides, of general formula $A_2B_2O_7$, have been the topic of much interest in recent years as they represent ideal systems for studying the effects of geometrical frustration [1]. Both the A and B sublattices form a network of corner-sharing tetrahedra such that it may not be possible to energetically satisfy all the magnetic interactions simultaneously. The resultant geometric frustration leads to the formation of exotic ground states. Much of the recent work has concentrated on local moment systems where novel properties such as cooperative paramagnetism [2], partial, noncollinear antiferromagnetic ordering [3,4], spin-freezing [5], and dipolar "spin-ice" behavior [6,7] have been observed. There has, however, been growing interest in the interplay between itinerant and local moments in geometrically frustrated systems. The metallic pyrochlore $Nd_2Mo_2O_7$ exhibits a large anomalous Hall effect which has been attributed to the Berry phase produced by spin chirality on the pyrochlore lattice [8], while the spinel, LiV_2O_4, may represent the first known transition metal heavy-fermion system, and evidence exists that the unusual properties of this material are related to geometrical frustration on the spinel lattice [9].

A vast body of work has been carried out on $3d$ and $4d$ transition metal pyrochlores. These are generally insulators and possess either a spin-glass-like or long-range ordered magnetic structure. In contrast, the $5d$ transition metal pyrochlores are mainly metallic, due to the extended nature of the $5d$ orbitals. The exception to this is $Cd_2Os_2O_7$ [10], where the $5d^3$ configuration of Os^{5+} results in a half-filled t_{2g} band and a metal-insulator transition at 226 K. Despite the large number of transition metal compounds which crystallize in the pyrochlore structure, superconductivity had not been observed until the recent discovery of bulk superconductivity in the $5d$ pyrochlore, $Cd_2Re_2O_7$ [11,12]. Recent investigations [11–15] have demonstrated the existence of two phase transitions in this compound. The first, occurring at $T \sim 200$ K, is a continuous structural transition accompanied by drastic changes in resistivity and magnetic susceptibility [14,15]. In addition, $Cd_2Re_2O_7$ has been shown to exhibit bulk superconductivity below a sample dependent $T_c \sim 1$ K [11–13]. Preliminary measurements in the superconducting state indicate that $Cd_2Re_2O_7$ is a type-II superconductor with H_{c1} less than 0.002 T and estimates of the upper critical field, H_{c2}, ranging from 0.2–1 T [11–13]. None of the measurements reported until now extend below 0.3 K ($T/T_c \sim 0.3$); hence, little can be concluded about the order parameter symmetry in this system. An exponential form of the specific heat as $T \rightarrow 0$ was speculated by Hanawa *et al.* [12], but they point out that measurements to lower temperatures are clearly needed. We report measurements [16] on $Cd_2Re_2O_7$ below 300 mK, temperatures necessary (for $T_c \sim 1$ K) to extract information about the superconducting order parameter symmetry. We have performed transverse-field (TF) and zero-field (ZF) muon spin rotation (μSR) measurements on single crystal samples of $Cd_2Re_2O_7$. The ZF-μSR measurements reveal very small internal magnetic fields characteristic of nuclear dipoles, indicating no significant electronic magnetism either above or below T_c. The TF-μSR results provide the first measurement of the internal field distribution in the vortex state in this material. In particular, temperature dependent studies from 20 mK to 4 K indicate a penetration depth which levels off as $T \to 0$, suggestive of a fully gapped Fermi surface with a rather large zero temperature value of the penetration depth, $\lambda(0) \sim 7500$ Å.

Muon spin rotation has proven to be a very effective probe in the study of superconductivity [17]. In particular, TF-μSR provides a measure of length scales associated with type-II superconductors, the penetration depth λ, and vortex core radius r_0 [17]. In a TF-μSR experiment, spin polarized muons, with polarization perpendicular to the applied magnetic field direction, are implanted in a sample at a location which is random on the length scale of the vortex lattice. The muon precesses at a rate proportional to the local magnetic field providing a measure of the local field distribution. The vortex lattice results in a spatially inhomogeneous field distribution and a resulting muon spin depolarization.

Early TF-μSR measurements assumed a Gaussian distribution of magnetic fields where the penetration depth is directly obtained from the Gaussian depolarization rate, $\sigma \sim 1/\lambda^2$. This approximation is reasonable for the case of polycrystalline samples but has been shown to be inadequate for single crystals [17]. In the single crystal case, a Ginzburg-Landau (GL) model for the magnetic field distribution has been developed. In this model, the size of the vortex core is determined by the applied magnetic field, H, and the GL coherence length normal to the applied field, $\xi_{\rm GL}$, while the penetration depth provides the length scale of the decay of the magnetic field away from the vortex core. The field distribution is calculated from the spatial distribution of the magnetic field [18],

$$B(\mathbf{r}) = \frac{\Phi_0}{S}(1 - b^4)\sum_{\mathbf{G}} e^{-i\mathbf{G}\cdot\mathbf{r}} \frac{uK_1(u)}{1 + \lambda^2 G^2}, \qquad (1)$$

where $u^2 = 2\xi_{\rm GL}^2 G^2(1 + b^4)[1 - 2b(1 - b^2)]$, $K_1(u)$ is a modified Bessel function, $\mathbf{G}$ is a reciprocal lattice vector of the vortex lattice, $b = H/H_{c2}$ is the reduced field, Φ_0 is the flux quantum, and S is the area of the reduced unit cell for a hexagonal vortex lattice.

Single crystals of $Cd_2Re_2O_7$ were grown using vapor-transport techniques as described elsewhere [13,19]. Three samples with an approximate surface area of (5×5) mm^2 each were mounted using low temperature grease with the cubic (100) direction parallel to the applied magnetic field direction. They were mounted on intrinsic GaAs to eliminate any precession signal at the background frequency [20] from muons which miss the sample and would otherwise land in the Ag sample holder. The samples were covered with 0.025 mm Ag foil which was bolted to the sample holder to ensure temperature uniformity. The TF and ZF-μSR measurements were performed in an Oxford Instruments dilution refrigerator on the M15 beam line at TRIUMF at temperatures from 20 mK up to 4 K.

Given the estimated critical field values, we selected a field of 0.007 T and the temperature dependence was measured by field cooling the sample to ensure a uniform flux line lattice. Figures 1(a) and 1(b) show typical μSR spectra in a transverse field of 0.007 T, for temperatures above and below T_c, respectively. Examination of this data clearly shows an enhanced depolarization rate on entering the superconducting state resulting from the inhomogeneous field distribution of the flux line lattice. This represents the first experimental observation of the vortex lattice in $Cd_2Re_2O_7$, providing clear evidence that this material is a type-II superconductor. Typical ZF spectra are shown in Fig. 1(c) for temperatures above and below T_c. The ZF relaxation is quite weak considering the large Re nuclear moments but, strangely, the form looks more exponential than Gaussian. One possibility is that there are multiple muon stopping sites with varying

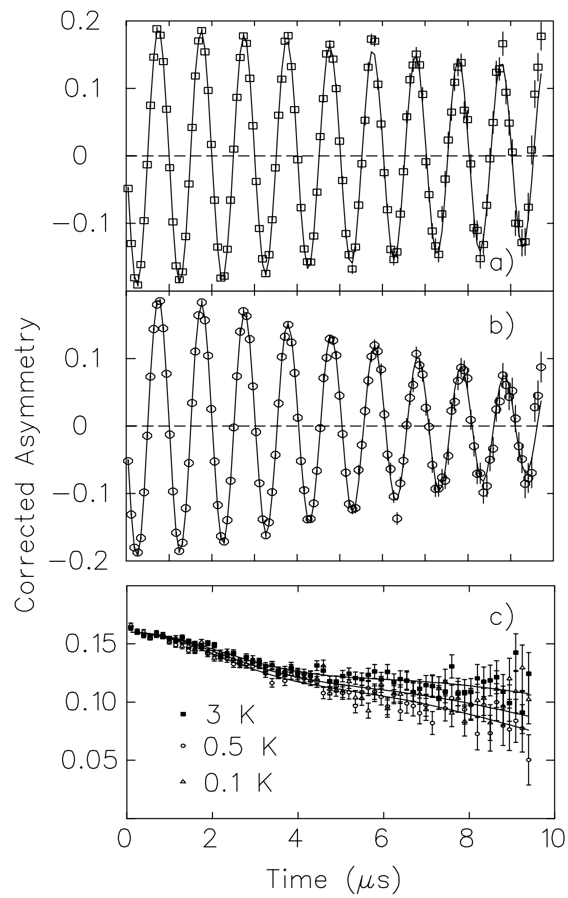

FIG. 1. Typical μSR spectra in $Cd_2Re_2O_7$ obtained in a transverse magnetic field of 0.007 T at temperatures of (a) $T = 1.5$ K (above T_c) and (b) $T = 100$ mK (below T_c). Typical ZF muon data are shown in (c) at several temperatures above and below T_c. The solid lines in (c) represents a fit to two Kubo-Toyabe functions as described in the text. Averaging over both muon sites, $\Delta = 0.114$, 0.131, and 0.144(7) μs^{-1} at 3, 0.5, and 0.1 K, respectively.

distance to the Re nuclear moments. The solid line in Fig. 1(c) assumes two muon sites, describing the muon spin relaxation by a Gaussian distribution of static internal fields at each site, a so-called Kubo-Toyabe function [21]. The second moment of the distribution is given by Δ^2/γ_μ^2, where γ_μ is the muon gyromagnetic ratio. Average values are given in Fig. 1. An exponential form for the relaxation could also be due to fluctuating electronic moments, as has recently been reported in [22]. However, in that case, an *increase* in TF linewidth with applied magnetic field would be anticipated. This is not observed in $Cd_2Re_2O_7$, as shown in Fig. 3.

There is also a slight increase in the apparent relaxation function below T_c. This is most likely not a real change in the internal field but rather the result of a small stray magnetic field along the muon polarization which is Meissner screened below T_c. In any event, the change in the relaxation function below T_c in ZF is much too small to explain the TF line broadening. Therefore, the increase in TF line broadening below T_c is attributed entirely to the vortex lattice.

The solid lines shown in Figs. 1(a) and 1(b) represent fits of the individual time spectra to a sample signal consisting of a Gaussian envelope with fixed asymmetry and a background signal with fixed linewidth and asymmetry. The background, from muons which miss the sample and land in the heat shields, was obtained independently by performing measurements with the sample removed. The resulting sample linewidth, σ, is shown in Fig. 2 as a function of temperature. As one can clearly see, the magnitude of the depolarization rate as $T \to 0$ is very small, saturating at a value of about 0.1 μs^{-1}. The residual linewidth from nuclear dipoles, taken from the data above T_c, is very small in $Cd_2Re_2O_7$ (about 0.03 μs^{-1}), allowing for clear observation of the line broadening associated with the flux line lattice.

As shown in Eq. (1), the field distribution depends on both the penetration depth and the GL coherence length. The GL coherence length can be obtained from the upper critical field, $\xi_{GL} = (\Phi_0/2\pi H_{c2})^{1/2}$, where Φ_0 is the flux quantum. As mentioned above, a range of values for H_{c2} have been reported and, consequently, to provide a self-consistent measurement of λ, the field dependence of the linewidth was measured. To account for any instrumental field dependence in the linewidth, measurements were made above the transition temperature (2 K) at each field after which the sample was field-cooled to 100 mK. The measured linewidth in the normal state was subtracted in quadrature from that observed at 100 mK and the results are plotted in Fig. 3. The corrected linewidth σ_{FC} decreases almost linearly with applied field, which is attributed to the linear increase in the volume taken up by the vortices. It approaches zero at a field of 5 kOe, our estimate of H_{c2} ($T \to 0$), and is consistent with measurements on other samples.

Using the corresponding $\xi_{GL} \sim 260$ Å, and Eq. (1), we obtain the temperature dependent penetration depth shown in Fig. 4. As expected from the small values of linewidth, at the base temperature we observe a rather large value of the penetration depth, $\lambda(0) \sim 7500$ Å. We note that this is significantly larger than most oxide superconductors where values ranging from 1000–2000 Å are typical [23,24].

As is clearly seen in Figs. 2 and 4, the linewidth and penetration depth, respectively, become temperature independent for $T \lesssim 0.4$ K, consistent with a fully gapped Fermi surface. Consequently, we conclude that the superconducting order parameter in $Cd_2Re_2O_7$ is consistent with a nodeless energy gap suggesting either *s*-wave symmetry or exotic pairing symmetries, such as *p*-wave which can also exhibit a fully gapped Fermi surface. For comparison, the solid line in Fig. 4 represents fits to the two fluid approximation,

$$\lambda^2(T) = \lambda^2(0)[1 - (T/T_c)^4]^{-1}, \quad (2)$$

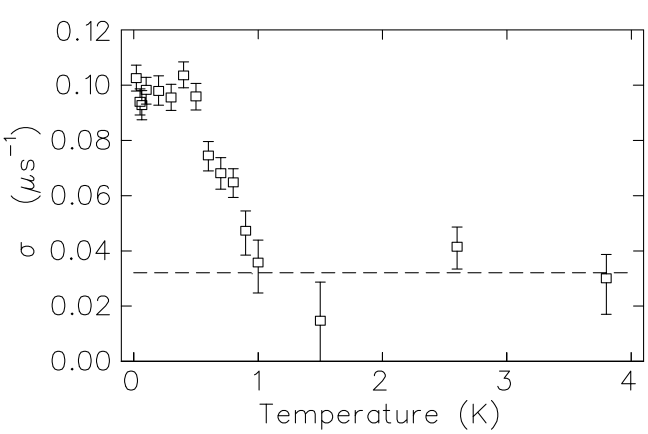

FIG. 2. Linewidth parameter σ as a function of temperature in a transverse magnetic field of 0.007 T applied along the (100) direction.

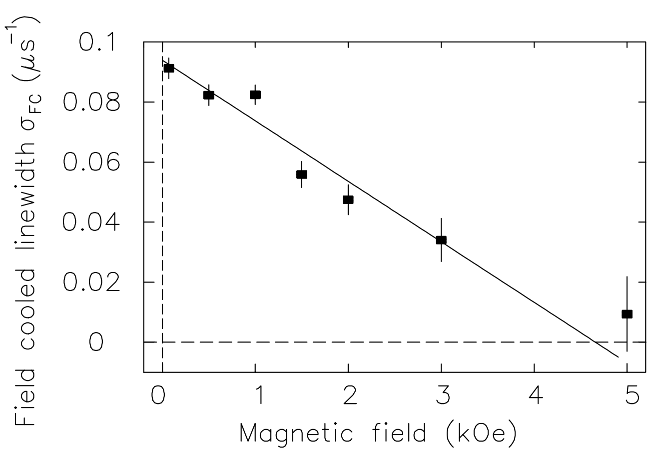

FIG. 3. Linewidth parameter σ_{FC} as a function of magnetic field applied along the (100) direction. The normal state contribution has been subtracted as described in the text. Measurements were taken at a temperature of 100 mK.

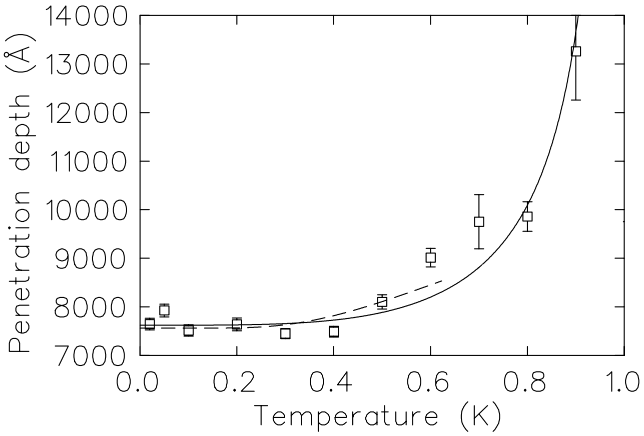

FIG. 4. Penetration depth as a function of temperature in a magnetic field of 0.007 T applied parallel to the (100) direction. The solid line is a fit to Eq. (2) and the dashed line is a fit to Eq. (3).

while the dashed line in Fig. 4 is a fit to the BCS temperature dependence,

$$\lambda(T) = \lambda(0)\left[1 + \sqrt{\frac{\pi\Delta_0}{2T}}\exp\left(\frac{-\Delta_0}{T}\right)\right], \quad (3)$$

where $\Delta_0 = 1.74(11)$ K.

The London penetration depth, λ, provides a direct measure of the ratio of superconducting carrier concentration to effective mass, n_s/m^*,

$$\frac{1}{\lambda^2} = \frac{4\pi n_s e^2}{m^* c^2}\left(1 + \frac{\xi_0}{l}\right)^{-1}, \quad (4)$$

where ξ_0 is the Pippard coherence length, and l is the mean-free path. There is considerable uncertainty in estimations of the mean-free path with reported values ranging from 200–700 Å [13,25], and it is unclear whether $Cd_2Re_2O_7$ is a superconductor in the clean or the dirty limit. If the material is in the clean limit ($\xi_0/l \ll 1$), the present results provide strong evidence for a fully gapped Fermi surface and we estimate a value of $n_s m_e/\mathrm{m}^* \sim 5.0 \times 10^{25}$ m^{-3} using Eq. (4) and the measured penetration depth. On the other hand, if $l \sim 200$ Å (i.e., the dirty limit), then we obtain $\xi_0 \sim 470$ Å and $n_s m_e/\mathrm{m}^* \sim 1.4 \times 10^{26}$ m^{-3}. Clearly, precise determination of the mean-free path for $Cd_2Re_2O_7$ is needed to allow accurate quantitative information to be extracted.

In conclusion, we have performed μSR studies of the superconducting state in the recently discovered pyrochlore superconductor, $Cd_2Re_2O_7$. Zero-field measurements indicate no significant magnetism in this superconductor, suggesting that magnetic frustration does not play a direct role in the superconductivity. Transverse-field measurements show that $Cd_2Re_2O_7$ is a type-II superconductor with an order parameter consistent with a fully gapped Fermi surface and a zero temperature penetration depth of $\sim$7500 Å. However, considering that the superconductor may be in the dirty limit, spectroscopic techniques which directly measure the density of states would be required to confirm this conclusion.

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