Quasiparticle Excitation in the Superconducting Pyrochlore $Cd_2Re_2O_7$ Probed by Muon Spin Rotation

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Abstract

The quasiparticle excitations in the mixed state of $Cd_2Re_2O_7$ have been studied by means of muon spin rotation/relaxation (μ SR). The temperature dependence of the magnetic penetration depth (λ) is consistent with a nearly isotropic superconducting order parameter, although a slight discrepancy which is dependent on the details in the analysis may be present. This is also supported by the relatively weak field dependence of λ .

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A class of metal oxides isostructural to mineral pyrochlore has been attracting considerable attention because they exhibit a wide variety of interesting physical properties. [1] The pyrochlore has a general formula of $A_2B_2O_7$, consisting of BO_6 octahedra and eightfold coordinated A cations, where A and B are transition metals and/or rare-earth elements. In particular, the B sublattice can be viewed as a three-dimensional network of corner-sharing tetrahedra, providing a testing ground for studying the role of geometrical frustration in systems which have local spins at B sites with antiferromagnetic (AFM) correlation. [2] Such systems are known to remain frustrated even when the exchange interaction is ferromagnetic (FM), provided that the spin correlation has local Ising anisotropy. [3] Recent studies have revealed a rich variety of phenomena seemingly related to the geometrical frustration, such as the occurrence of a spin-glass (SG) phase in $R_2Mo_2O_7$ with R=Y, Tb and Dy [4,5], the unusual behavior of ordinary and anomalous Hall coefficients in the same compound with R=Nd, Sm and Gd [6,7], and the "spin-ice" phase in $R_2Ti_2O_7$ with R=Dy and Ho. [3,8]

Although metallic pyrochlores comprise a minority subgroup of the pyrochlore family, they consist of distinct members such as $Tl_2Mn_2O_7$, which exhibits colossal magnetoresistance. [9,10] Moreover, the recently revealed superconductivity in $Cd_2Re_2O_7$, a 5d transition metal pyrochlore [11,12], demonstrates that the pyrochlores provide a fertile field for electronic correlation adjacent to the perovskite compounds. In this context, it is noteworthy that LiV_2O_4 , a cubic spinel compound in which the V sublattice is isostructural to the B sublattice in pyrochlore, behaves similarly to a heavy fermion metal. [13]

It is reported that $Cd_2Re_2O_7$ falls into the bulk superconducting state below $T_c \simeq 1 \sim 2$ K, as confirmed by a large jump of specific heat ΔC_c as well as large diamagnetism due to the Meissner effect associated with the occurrence of zero resistivity. [11] The dc magnetization curve indicates that the superconductivity is of type II with the upper critical field close to 0.29 T at 0 K. [14] The ratio $\Delta C_c/\gamma T_c$ (with γ being the Sommerfeld constant) is reported to be 1.15, which is smaller than the predicted value of 1.43 for isotropic BCS superconductors. Unfortunately, these measurements were performed only above 0.4 K and are thus inconclusive in determining the detailed characteristics of superconductivity in

 $Cd_2Re_2O_7$. In this Letter, we report on the quasiparticle excitations in the mixed state of $Cd_2Re_2O_7$ studied by muon spin rotation/relaxation (μ SR). The magnetic penetration depth λ , which reflects the population of normal electrons ("quasiparticles") in the superconductive state, is determined microscopically by measuring the muon spin relaxation due to the spatial inhomogeneity of magnetic induction in the flux line lattice (FLL). We show that the temperature dependence of λ is more or less consistent with the prediction based on the BCS superconductors with an isotropic gap, although a slight discrepancy is suggested by the detailed analysis.

The μ SR experiments on both single-crystal and polycrystalline Cd₂Re₂O₇ were performed on the M15 beamline at the TRIUMF muon facility which provides a beam of nearly 100% spin-polarized positive muons of momentum 28.6 MeV/c. The specimen was mounted on the coldfinger of a 3 He- 4 He dilution refrigerator and cooled from a temperature above T_c after setting the magnetic field at every field point (i.e., field-cooling) to eliminate the effect of flux pinning. The field and temperature scan data were obtained at T=0.2 K and at H=0.1 T, respectively. Muons were implanted into the specimen (measuring about 10 mm×10 mm and 1 mm thick) after being passed through a beam collimator. The initial muon spin polarization was perpendicular to the magnetic field H and thus to the FLL in the superconducting state.

Since the muons stop randomly along the length scale of the FLL, the muon spin precession signal $\hat{P}(t)$ provides a random sampling of the internal field distribution $B(\mathbf{r})$,

$$\hat{P}(t) \equiv P_x(t) + iP_y(t) = \int_{-\infty}^{\infty} n(B) \exp(i\gamma_{\mu}Bt) dB, \qquad (0.1)$$

$$n(B) = \frac{d\mathbf{r}}{dB},\tag{0.2}$$

where γ_{μ} is the muon gyromagnetic ratio (=2 π ×135.53 MHz/T), and n(B) is the spectral density for muon precession determined by the local field distribution. These equations indicate that the real amplitude of the Fourier-transformed muon precession signal corresponds to the local field distribution n(B). The local field distribution can be approximated as the sum of magnetic induction from isolated vortices in the London model to yield

$$B(\mathbf{r}) = B_0 \sum_{\mathbf{K}} \frac{e^{-i\mathbf{K}\cdot\mathbf{r}}e^{-K^2\xi_v^2}}{1 + K^2\lambda^2 + O(K_x^2, K_y^2)},$$
(0.3)

where **K** is a translation of the vortex reciprocal lattice, B_0 ($\simeq H$) is the average internal field, λ is the London penetration depth, and ξ_v is the cutoff parameter. The term $O(K_x^2, K_y^2)$ denotes the nonlocal effect in which the electromagnetic response kernel $Q(\mathbf{K})$ generating the supercurrent around the vortex depends on **K**. While this term is eliminated in the conventional BCS superconductors with isotropic s-wave pairing, it becomes important for the moe complex order parameters such as anisotropic s-wave or d-wave (e.g., $d_{x^2-y^2}$). The London penetration depth in the FLL state is related to the second moment $\langle \Delta B^2 \rangle = \langle (B(\mathbf{r}) - B_0)^2 \rangle$ of the field distribution reflected in the μ SR line shape (where $\langle \rangle$ means the spatial average). In polycrystalline samples, a Gaussian distribution of local fields is a good approximation, where

$$\hat{P}(t) \simeq \exp(-\sigma^2 t^2/2) \exp(i\gamma_\mu H t), \tag{0.4}$$

$$\sigma = \gamma_{\mu} \sqrt{\langle \Delta B^2 \rangle}. \tag{0.5}$$

For the case of ideal triangular FLL with isotropic effective carrier mass m^* and a cutoff $K \approx 1.4/\xi_v$ provided by the numerical solution of the Ginsburg-Landau theory, the London penetration depth λ (with $O(K_x^2, K_y^2) = 0$) can be deduced from σ using the following relation [15–17],

$$\sigma \left[\mu s^{-1}\right] = 4.83 \times 10^4 (1 - h) \lambda^{-2} \text{ [nm]},$$
(0.6)

where $h = H/H_{c2}$. While the above form is valid for h < 0.25 or h > 0.7, a more useful approximation valid for an arbitrary field is [17]

$$\sigma \left[\mu s^{-1}\right] = 4.83 \times 10^{4} (1 - h) [1 + 3.9(1 - h)^{2}]^{1/2} \lambda^{-2} \text{ [nm]}.$$
 (0.7)

In both cases, λ is related to the superconducting carrier density n_s as

$$\lambda^2 = \frac{m^* c^2}{4\pi n_s e^2},\tag{0.8}$$

indicating that λ is enhanced upon the reduction of n_s due to the quasiparticle excitations. For simplicity, we adopt eq. (0.6) for the following analysis.

In a preliminary analysis, we found that the spin relaxation rate due to the FLL formation is less than $0.1 \ \mu s^{-1}$ which is typically of the same order of magnitude as that due to static random local fields from nuclear magnetic moments. This means that the additional relaxation due to 111,113 Cd and 185,187 Re nuclear moments must be considered for the proper estimation of λ in Cd₂Re₂O₇. To this end, the following equation was used in the actual fitting analysis of the time spectra,

$$\hat{P}(t) = \exp[-(\sigma_0 t)^{\nu} + \sigma^2 t^2 / 2)] \exp(i\gamma_{\mu} H t), \tag{0.9}$$

where σ_0 is the relaxation rate due to the nuclear moments and ν is the power of relaxation. The parameters σ_0 and ν were evaluated by fitting the time spectra above T_c with σ set to zero, yielding $\sigma_0 \simeq 0.057(1) \ \mu\text{s}^{-1}$ and $\nu \simeq 1.0$ in the polycrystalline specimen at H = 0.1 T. Then, σ due to the formation of FLL below T_c was deduced by analyzing data with σ_0 and ν being fixed to the above values. A similar analysis was performed for the data obtained for the single crystals.

Figure 1(a) shows the temperature dependence of σ in Cd₂Re₂O₇ at H=0.1 T. Upon the onset of FLL formation, σ exhibits a gradual increase with decreasing temperature just below $T_c(0.1 \text{ T}) \sim 0.7 \text{ K}$ at this field. According to the empirical two-fluid model approximately valid for conventional BCS superconductors, we have

$$\lambda(t) = \lambda(0) \frac{1}{\sqrt{1 - t^4}},\tag{0.10}$$

which leads to

$$\sigma(t) = \sigma(0)(1 - t^4), \tag{0.11}$$

where $t \equiv T/T_c(0.1 \text{ T})$. The fitting analysis by the same formula with an arbitrary power,

$$\sigma(t) = \sigma(0)(1 - t^{\beta}), \tag{0.12}$$

with T_c as a free parameter yields $\beta = 2.8(5)$ and $T_c = 0.65(3)$ K. We also found that $T_c = 0.62(1)$ K when $\beta = 4$ is assumed, which is slightly lower than the value $T_c \simeq 0.7$ K estimated from the specific heat measurement for this field. Although the difference is not obvious between these cases (solid curve for $\beta = 2.8$ and the dashed curve for $\beta = 4$) in Fig. 1(a), we note that the reduced χ^2 for the former is almost two times smaller (better) than the latter. Taking $H_{c2} = 0.29$ T, the penetration depth extrapolated to T = 0 ($\lambda(0, 0.1 \text{ T})$) is 700(8) nm, leading to the Ginsburg-Landau parameter $\kappa \simeq 21$ with $\xi \simeq 34$ nm estimated from $H_{c2}(0)$. Thus, it is concluded that $Cd_2Re_2O_7$ is a typical type II superconductor with large λ , which is consistent with the results of magnetization measurement. [11]

The finding that $\beta \simeq 2.8$ may indicate that the deviation $\Delta \lambda = \lambda(t) - \lambda(0) \propto T^{\beta}$ exhibits a tendency predicted for the case of line nodes (*d*-wave pairing) with some disorder (i.e., dirty limit), where $\Delta \lambda \propto T^2$. [18] Such a temperature dependence has actually been observed in high- T_c (YBCO) cuprates. [19,20] Compared with the case of isotropic gap $\Delta_{\hat{k}} = \Delta_0$, the quasiparticle excitations are enhanced along nodes ($|\Delta_{\hat{k}}| = 0$) to reduce average n_s , leading to the enhancement of λ . However, the result of the fitting analysis also suggests that the discrepancy may be attributed to experimental uncertainty including the precise value of T_c . Thus, we are led to conclude that the order parameter in Cd₂Re₂O₇ is mostly isotropic with a possibility of residual weak anisotropy as suggested by the slightly reduced value of β .

As shown in Fig. 2(a), σ decreases with increasing external field, where the general field dependence is determined by the increasing contribution of normal vortex cores and the stronger overlap of field distribution around the cores which are described by the term (1-h) in eq.(0.6). The fitting analysis by eq.(0.6) with H_{c2} and $\lambda(H=0)$ as free parameters yields $H_{c2} = 0.37(5)$ T and $\lambda(H=0) = 796(12)$ nm at 0.2 K. Since the deduced value of H_{c2} is consistent with that obtained from the magnetization measurement ($\simeq 0.29$ T), we can conclude that the observed field dependence of λ is mostly due to the vortex core/overlap effect, except for the fields below ~ 0.06 T where a slightly steeper reduction of σ is suggested. The penetration depth deduced for each field using eq.(0.6) with $H_{c2} = 0.37$ T is plotted in

Fig. 2(b). In order to evaluate the relative strength of the pair-breaking effect, it is useful to introduce a dimensionless parameter η to describe the field dependence of λ with the following simple linear relation,

$$\lambda = \lambda(0.2K, h)[1 + \eta h], \tag{0.13}$$

where $h \equiv H/H_{c2}(0.2\text{K})$ with $H_{c2}(0.2\text{K}) = 0.37$ T. From the analysis of data in Fig. 2(b) by eq. (0.13), we find that $\eta = 0.38(14)$ with $\lambda(0.2\text{K}, 0) = 741(5)$ nm for $0 \le H \le 0.06$ T.

In general, the field dependence of λ is enhanced by two different mechanisms, i.e., the nonlinear effect in the semiclassical Doppler shift of the quasiparticle energy levels due to the supercurrent around the vortex cores, and the nonlocal effect which further modifies the quasiparticle excitation spectrum in the momentum space. In particular, the nonlocal effect is important in the system with line nodes because the coherence length is inversely proportional to the order parameter, such that $\xi_0(\hat{k}) = \hbar v_F/\pi \Delta_{\hat{k}}$. The divergence of ξ_0 along the nodal directions $|\Delta_{\hat{k}}| = 0$ means that the response of quasiparticles near the nodes is highly nonlocal.

It is predicted that $\eta \ll 1$ for the isotropic s-wave pairing because the finite gap prevents the shifted levels of quasiparticle excitations from being occupied at low temperatures. This is supported, for example, by the recent observation in CeRu₂, in which $\eta \simeq 0$ over the field region $0 \le h \le 0.5$ where the system behaves more or less as a conventional BCS superconductor with isotropic s-wave pairing. [21] On the other hand, stronger field dependence is predicted for the case of d-wave or anisotropic s-wave pairing due to the excess population of quasiparticles in the region where $|\Delta_{\hat{k}}|$ is small or zero. Typical examples for the d-wave pairing are those of high- T_c cuprates in which η is reported to be 5~6.6 for YBCO. [22] Meanwhile, in the case of an s-wave superconductor YNi₂B₂C in which strong anisotropy for $\Delta_{\hat{k}}$ is suggested experimentally [23], $\eta \simeq 1$ is reported from a μ SR study. [24] The comparison of these earlier results with our result suggests that the anisotropy of the order parameter in Cd₂Re₂O₇ is considerably smaller than YNi₂B₂C.

Finally, we comment that the recent observation of a clear coherence peak below T_c in

the 187 Re NQR measurement [25] does not necessarily indicate the absence of anisotropy in the order parameter. In the case of the anisotropic energy gap, the magnitude of the coherence peak depends on the mean free path l. The coherence peak is enhanced in a certain condition of l where quasiparticles probe only a limited region of the Fermi surface. Thus, while the actual value of l is difficult to estimate because of the semimetallic character of this compound, the NQR result does not completely rule out the presence of anisotropy in general.

In summary, we have investigated the quasiparticle excitations in the mixed state of $Cd_2Re_2O_7$ by μSR . The temperature and field dependence of the London penetration depth indicates that the basic feature is consistent with the isotropic order parameter for BCS s-wave pairing, although there remains a certain subtlety suggested by the small deviation from the theoretical prediction which may be better understood by considering a weak anisotropy.

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FIGURES

- FIG. 1. Temperature dependence of a) the muon spin relaxation rate σ due to flux line lattice and b) the magnetic penetration depth λ , where solid circles are the data from the polycrystalline specimen and open triangles are those from single crystals. Solid curves are results of fitting by a relation $\sigma \propto 1/\lambda \propto 1 (T/T_c)^{\beta}$ with β and T_c being free parameters. Dashed curves are obtained when $\beta = 4$ with T_c as a free parameter.
- FIG. 2. Magnetic field dependence of a) the muon spin relaxation rate σ due to flux line lattice and b) the magnetic penetration depth λ . The solid line in a) is a result of fitting by eq.(0.6) with H_{c2} being a free parameter. Dashed lines are obtained by fitting data with a relation $\lambda \propto 1 + \eta(H/H_{c2})$ only for $0 \le H \le 0.06$ T.



