Geometrically frustrated spin systems have been attracting much attention. Since a three-dimensional network of corner-sharing tetrahedra, a so-called pyrochlore lattice, exhibits magnetic frustration in the case of antiferromagnetic exchange interaction between neighboring magnetic ions, no magnetic ordering appears, leading to the spin liquid ground state.\(^1\) It is noteworthy that electron transport under such strong magnetic correlations exhibits interesting properties, since the electron correlation also has an implicit relationship with the charge and/or orbital degrees of freedom. In particular, in the case of a mixed valence state, i.e., half-integer numbers of \(d\)-electrons per magnetic site, it is suggested that charge/orbital fluctuation plays an essential role. For instance, the heavy-fermion-like behavior of \(\text{LiV}_2\text{O}_4\) has been reported,\(^2\) in which each \(\text{V}^{3+}\) ion forming a pyrochlore lattice accommodates 1.5\(d\)-electrons. It has been shown that the electronic state exhibits strong correlation under geometrical frustration.\(^3\) This compound is one of only two spinel oxides showing metallic conductivity without any structural transition even at low temperatures. Another example is \(\text{Li}_2\text{Ti}_2\text{O}_7\) having 0.5\(d\)-electrons per \(\text{Ti}^{3+}\) ion, which is known to be a superconductor with \(T_c \approx 13\,\text{K}\).\(^4\) It has turned out, on the other hand, that many of the metallic compounds having magnetic frustration show the metal–insulator (MI) transition upon cooling. As found in the cases of \(\text{AlV}_2\text{O}_4\)\(^5\) and \(\text{CuIr}_2\text{S}_4\),\(^6\) some compounds exhibit charge ordering and an associated structural change that removes the geometrical degeneracy. However, in other cases, e.g., \(\text{Cd}_2\text{Os}_2\text{O}_7\),\(^7,8\) no structural transition is seen, suggesting that the electronic properties are predominantly controlled by magnetic frustration.

Recently, Yonezawa et al. discovered that \(\text{KO}_2\text{Os}_6\), which has a \(\beta\)-pyrochlore structure, shows bulk superconductivity below \(T_c \approx 9.6\,\text{K}\).\(^9\) Moreover, a series of \(\beta\)-pyrochlore superconductors, \(\text{RbOs}_2\text{O}_6\) (\(T_c \approx 6.3\,\text{K}\)) and \(\text{CsOs}_2\text{O}_6\) (\(T_c \approx 3.3\,\text{K}\)), was subsequently found.\(^10,11\) Until these discoveries, \(\text{Cd}_2\text{Re}_2\text{O}_7\) was the only pyrochlore oxide known to show superconductivity (\(T_c \approx 1\,\text{K}\)).\(^12\)–\(^14\) As in the case of \(\text{Cd}_2\text{Re}_2\text{O}_7\), a \(5d\) transition metal forms a pyrochlore lattice in \(\beta\)-pyrochlore superconductors. Nevertheless, it is notable that the formal oxidation state of the \(\text{Os}^{5+}\) ion is 5.5\(+\) (5\(d^5\)), which is specific to the \(\beta\)-pyrochlore structure, while that of the \(\text{Re}^{5+}\) ion in \(\text{Cd}_2\text{Re}_2\text{O}_7\) is \(5+\) (5\(d^2\)). \(\text{Os}_2\text{O}_7\) having Os\(5+\) (5\(d^5\)) exhibits the MI transition at \(\approx 230\,\text{K}\),\(^15\) suggesting the presence of strong correlation among \(d\)-electrons even in the case of \(\text{KO}_2\text{Os}_6\).

Furthermore, the geometrical frustration in \(\text{KO}_2\text{Os}_6\) naturally gives rise to the idea of superconductor based on the spin liquid state. It is noteworthy that \(\text{Na}_2\text{Co}_2\text{O}_7\cdot\text{yH}_2\text{O}\), having a two-dimensional triangular lattice of \(\text{CoO}_6\), has been recently identified as a superconductor with \(T_c \approx 5\,\text{K}\).\(^15\) The triangular lattice is well known as a stage of geometrical frustration. Moreover, it is suggested from chemical titration and photoemission studies that cobalt ions are in a mixed valence state (Co\(^{3+}\)\(^{4+}\)).\(^16\)

It is interesting to see whether or not the superconducting state of \(\text{KO}_2\text{Os}_6\) is similar to that of \(\text{Cd}_2\text{Re}_2\text{O}_7\), because the resistivity in the normal state of \(\text{KO}_2\text{Os}_6\) shows an anomalous broad shoulder with varying temperature, and no phase transition down to \(\approx 4\,\text{K}\) has been confirmed by X-ray diffractometry,\(^17\) which is in marked contrast to the case of \(\text{Cd}_2\text{Re}_2\text{O}_7\).\(^18\) In this letter, we report the results of a muon spin rotation (\(\mu\text{SR}\)) experiment on \(\text{KO}_2\text{Os}_6\) under several transverse fields (TF) up to 6\(T\). The observed temperature dependence of the magnetic penetration depth \(\lambda_{\text{eff}}\) deduced from the TF–\(\mu\text{SR}\) measurement cannot be explained by the empirical two-fluid model. Furthermore, the field dependence of \(\lambda_{\text{eff}}\) strongly suggests the existence of low-lying quasiparticle excitations, which are attributed to either an anisotropic superconducting order parameter or a multigap structure in \(\text{KO}_2\text{Os}_6\).

The \(\mu\text{SR}\) experiment was conducted at the M15 beamline
of TRIUMF, Canada. Time-dependent muon polarization under a transverse field was measured using a high-time-resolution \(\mu\)SR spectrometer furnished with a superconducting magnet. For each \(\mu\)SR measurement, the magnetic field was applied at 20 K, which is well above \(T_c\), and subsequently the specimen was cooled to the lowest temperature of 2 K. The measured KO\(_2\)O\(_6\) sample was prepared as a pellet of polycrystalline powder in a square shape of 8 \(\times\) 8 mm\(^2\). Positive muons with a beam momentum of 29 MeV/c, whose polarizations were normal to the beam axis, were implanted. The \(\mu^+\rightarrow e^+\) decay asymmetry and associated Larmor precession with a frequency \(\omega \equiv \gamma_\mu B\) (where \(\gamma_\mu = 2\pi \times 135.54\) MHz/T is the muon gyromagnetic ratio) were measured to deduce the local field distribution. The decay events in which muons missed the sample were eliminated with an anticoincidence electronics logic circuit, resulting in quite low background positron spectra.

It is well known that type-II superconductors show the flux-line lattice (FLL) state at a field \(B_{c1} < B < B_{c2}\), leading to the spatial inhomogeneity of magnetic induction. Since the implanted muons occupy random positions over the length scale of FLL, i.e., \(\lambda_{\text{eff}}\), the observed precessing signal \(\hat{P}(t)\) is a spatial sum of the internal field, given by

\[
\hat{P}(t) = P_c(t) + i P_s(t) = \int_{-\infty}^{\infty} n(B) \exp(i\gamma_\mu B t) dB,
\]

where \(n(B)\) is the spectral density for muon precession determined by the local field distribution. In polycrystalline samples, the Gaussian distribution of local fields is a good approximation, namely

\[
\hat{P}(t) \approx \exp(-\sigma^2 t^2/2) \exp(i\gamma_\mu B t),
\]

\[
\sigma = \gamma_\mu \sqrt{\langle \Delta B^2 \rangle},
\]

where \(\langle \Delta B^2 \rangle\) is the average of the second moment of the field distribution \(\langle B(r) - B_0 \rangle^2\). By simulating the distribution of \(\lambda_{\text{eff}}\), we confirmed that the above assumption is reasonable, where the distribution is probably due to randomness of FLL coming from various imperfections in a real system.\(^{19}\) The field variation for an ideal triangular FLL is approximately estimated:\(^{20}\)

\[
\sigma (\mu\text{s}^{-1}) \approx 4.83 \times 10^4 (1 - b) \times [1 + 3.9(1 - b)^3]^{1/2} \lambda_{\text{eff}}^2 \text{ (nm)},
\]

where \(b \equiv B/B_{c2}\). Thus, \(\lambda_{\text{eff}}\) can be deduced directly from the measured relaxation rate \(\sigma^{21}\).

In Fig. 1, we show the fast Fourier transform (FFT) of the obtained TF-\(\mu\)SR time spectra at different temperatures. The line shape exhibits apparent broadening and a shift of the peak to lower frequencies below \(T_c \approx 9\) K, which is ascribed to the formation of the FLL state. On the other hand, there exists a relatively narrow peak at around the central frequency corresponding to the applied field, clearly suggesting that a nonsuperconducting (normal) portion was involved in the measured sample. The peak slightly moves to higher frequency below \(T_c\). Such behavior is often seen in the case of polycrystalline superconductors, e.g., MgB\(_2\)\(^{22}\), due to the local demagnetization of grains. The volume fraction of the normal part is estimated by fitting analysis to be about 20\%, which is in line with that found by the bulk susceptibility measurement.\(^9\) After the \(\mu\)SR measurement, it turned out that the employed sample involves a moderate amount of KO\(_2\)O\(_4\), which is an insulator. The analysis of the \(\mu\)SR experiment was carried out on the time-differential data using the sum of the two precessing parts given by eq. (3). Thus, we can obtain the field distribution solely ascribed to the superconductivity of KO\(_2\)O\(_6\). The temperature dependence of the muon spin relaxation rate of the superconducting part is shown in Fig. 2. In general, the London penetration depth \(\lambda_L\) is given by the following relation

\[
\lambda_L^2 = \frac{m^* c^2}{4\pi n_s e^2},
\]

where \(m^*\) is the effective mass and \(n_s\) is the superconducting carrier density. According to the Gorter–Casimir two-fluid model, the superconducting carrier density \(n_s\) is proportional to \(1 - (T/T_c)^4\). Thus, we have \(\lambda_L \propto 1/\sqrt{1 - (T/T_c)^4}\), leading to \(\sigma \propto 1 - (T/T_c)^4\). As shown in Fig. 2, our data exhibit significant deviation from this relation. The fitting analysis was then performed with an arbitrary power

\[
\sigma = \sigma_{T=0} \left[ 1 - \left( \frac{T}{T_c} \right)^\beta \right],
\]

with \(T_c\) as a free parameter. Here, the contribution from the nuclear magnetic moments such as \(^{39}\)K and \(^{189}\)O seen at \(T > T_c\) was subtracted by a fixed amount prior to the fitting analysis. While the best fitting is obtained when \(\beta = 2.39(7)\) and \(T_c = 8.91(3)\), the observed temperature dependence is
are shown in Fig. 2 together with the fitting result by the approximately linear tendency is seen below \( C_2 \) curvature with respect to the applied field. In particular, an excess quasiparticles due to the thermal activation at a finite temperature exist in the vicinity of gap minimum even in the case of anisotropic pairing symmetry. Note that the particle excitation in the case of anisotropic pairing symmetry is field dependent, showing a slight negative divergence at the nodes, because the superconducting carrier density decreases due to the low-lying quasiparticle excitations. The results of KOs

\[ KO_2S_2O_6 (B_0 = 2T) \]

are normalized field, as shown in Fig. 3(b). Apparently, the superconducting carrier density \( n \) is field dependent, showing a slight negative curvature with respect to the applied field. In particular, an approximately linear tendency is seen below \( 3T \).

In order to examine the low-lying quasiparticle excitations of \( KO_2S_2O_6 \), we measured the field dependence of \( \lambda_{\text{eff}} \) at the lowest temperature of 2 K. In Fig. 3(a), transverse relaxation rates at various fields are shown. Although \( \lambda_{\text{eff}} \) can be evaluated using eq. (5), the exact value of \( B_{C2} \) in the examined specimen is still unknown. Assuming that \( B_{C2} \approx 40T \), we deduced \( \lambda_{\text{eff}} \) versus the corresponding normalized field, as shown in Fig. 3(b). Apparently, the obtained \( \lambda_{\text{eff}} \) is field dependent, showing a slight negative curvature with respect to the applied field. In particular, an approximately linear tendency is seen below \( 3T \).

In general, \( \lambda_{\text{L}} \) is enhanced under a sufficiently high field, because the superconducting carrier density \( n \) decreases due to pair-breaking interactions such as the Zeeman interaction. However, the magnitude of this effect is small in conventional superconductors with isotropic \( s \)-wave pairing. On the other hand, a strong enhancement of \( \lambda_{\text{eff}} \) with external field is expected in the case of superconductors with the anisotropic order parameter due to the combined contribution of (i) nonlinear and (ii) nonlocal effects. More specifically, supercurrent \( v_\perp \) induced by the magnetic field causes a semiclassical Doppler shift of the quasiparticle energy levels, which is proportional to \( v_\perp \cdot v_F \), where \( v_F \) is the Fermi velocity. This gives rise to a nonlinear response of the shielding current to the field due to the “backflow”. More importantly, the Doppler shift enhances the quasiparticle excitation in the case of anisotropic pairing symmetry due to the low-lying quasiparticle excitations. Note that the excess quasiparticles due to the thermal activation at a finite temperature exist in the vicinity of gap minimum even in the case of anisotropic \( s \)-wave pairing. No such enhancement is expected in the case of isotropic \( s \)-wave superconductors as long as the gap energy is greater than the energy of the Doppler-shifted quasiparticles. Thus, the present result clearly exhibits the existence of low-lying quasiparticle excitations associating with the superconducting energy gap smaller than \( 2K (\approx 0.2 meV) \).

When the superconducting energy gap has nodes at the Fermi surface, the field dependence of \( \lambda_{\text{eff}} \) is further modified by the nonlocal effect. The coherence length \( \xi_\perp \) diverges at the nodes, because \( \xi_\perp \) is inversely proportional to the gap size \( \xi_\parallel = |v_F|/\pi \Delta_F \), where \( \Delta_F \) is the angular-dependent energy gap at the Fermi surface, resulting in the reformation of the FLL. As shown in Fig. 3(b), \( \lambda_{\text{eff}} \) tends to exhibit saturating behavior at higher fields. It is interesting to point out that a similar tendency has been discussed in the case of \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) by considering nonlinear and nonlocal effects, both theoretically and experimentally. In any case, we would like to emphasize that the field-dependent behavior of \( \lambda_{\text{eff}} \) is characterized by a small energy scale less than \( 0.2 \) meV.

To evaluate the strength of the pair-breaking effect, we performed a fitting using a simple linear relation

\[ \lambda_{\text{eff}} = \lambda_0 (1 + \eta B). \]

From the analysis of data below \( 3T \), we obtain \( \lambda_0 = 270 \) nm and \( \eta = 2.58 (T/T_c \approx 0.22) \), which is shown as the solid line in Fig. 3(b). We stress that \( \lambda_{\text{eff}} \) is independent of the applied field in the case of isotropic \( s \)-wave pairing symmetry. In \( \text{V}_3\text{Si} \), which is a typical \( s \)-wave superconductor, \( \eta \approx 0 \) is confirmed at lower fields (\( B/B_{C2} < 0.5 \)). This is in marked
In contrast to the present case. Besides this, it has turned out that MgB$_2$ exhibits $\eta = 1.27$ at 0.26 $T_c$. In this case, it is suggested that the presence of two superconducting energy gaps, one being relatively small ($\sim$1 meV), is responsible for the field dependence of $\Delta_{eff}$ observed at a finite temperature. In this sense, we cannot exclude the multiple-gap scenario in KO$_2$O$_6$, since the measurement was performed at a finite temperature of 2 K. The present result suggests the presence of a small energy gap having a magnitude of less than $\sim$0.2 meV in the multiple-gap scenario. It is noted, however, that distinguishing between the anistropic-gap case and the multiple-gap case is difficult with our present experimental data. In order to clarify this, a measurement at a further low temperature is planned. On the other hand, the finite value of $\eta = 2.58$ in KO$_2$O$_6$ is obviously different from that reported in the case of Cd$_2$Re$_2$O$_7$. There exists a crucial effect on the pairing mechanism. In contrast, the absence of structural transitions in KO$_2$O$_6$ suggests that the geometrical degeneracy remains even at low temperatures, leading to a spin liquid state, in which a dynamical magnetic correlation exists. One may expect that the Cooper pair is mediated by magnetic fluctuation under such circumstances.

In summary, we have investigated the quasiparticle excitation in KO$_2$O$_6$ by measuring the magnetic penetration depth $\lambda_{eff}$. The temperature dependence of $\lambda_{eff}$ exhibits a significant deviation from that expected for the empirical two-fluid model. Moreover, it was seen that $\lambda_{eff}$ increases markedly with applied external field up to 6 T, suggesting the presence of nonlinear and nonlocal effects. We consider this to be evidence that the superconducting order parameter realized in KO$_2$O$_6$ has a strong anisotropy or multigapped structure with a small gap energy.

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16) H. Sakurai: private communication.