Magnetic ordering in spin-orbit Mott insulator Ba$_2$IrO$_4$ probed by μSR

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Abstract. Magnetic properties in the novel spin-orbit Mott insulator Ba$_2$IrO$_4$ were studied using muon spin rotation (μSR) technique. Zero-field μSR experiments revealed that Ba$_2$IrO$_4$ shows an antiferromagnetic transition at $T_N \sim 240$ K without any spontaneous magnetization. The most stable μ$^+$ site was determined by the electrostatic (Madelung) potential calculation. The effective magnetic moment of the iridium ions ($|\mu|$) in the antiferromagnetic ordered state was calculated using a dipolar-field model, with an internal field obtained by μSR experiments. The magnetic moment is significantly reduced ($|\mu| \sim 0.34 \mu_B$) due to a low-dimensional quantum spin fluctuation with a large intra-plane correlation. The magnetic ground state of the spin-orbit Mott insulator Ba$_2$IrO$_4$ is quite similar to those in parent materials of high-$T_c$ cuprate superconductors such as La$_2$CuO$_4$.

1. Introduction

The recent findings of the novel Mott insulating state in Sr$_2$IrO$_4$ [1] have attracted particular interest of scientists, as a spin-orbit (SO) coupling induced Mott insulator. This novel electronic state is driven by large SO interaction and modest on-site Coulomb interaction in the 5$d$ electron system. The large SO interaction splits the iridium 5$d$ $t_{2g}$ orbital into a half-filled $J_{\text{eff}} = 1/2$ doublet and a $J_{\text{eff}} = 3/2$ quadruplet. The on-site Coulomb interaction further splits the $J_{\text{eff}} = 1/2$ band into the upper and lower Hubbard bands, yielding the SO Mott insulating state. The SO $J_{\text{eff}} = 1/2$ Mott state seems like ground states in parent materials of high-$T_c$ cuprate superconductors. One may therefore expect that unconventional superconductivity occurs in carrier-doped SO Mott insulators.

Recently, we succeeded in synthesizing a novel SO Mott insulator Ba$_2$IrO$_4$ under high pressure [2]. Ba$_2$IrO$_4$ is isostructural to Sr$_2$IrO$_4$, but has flat IrO$_2$ square planar lattices with straight Ir-O-Ir bonds. As a result, Ba$_2$IrO$_4$ has a genuine (non-canted) antiferromagnetic ordered state without a spontaneous magnetic moment below $T_N$ (~240 K) due to absence of Dzyaloshinsky-Moriya interaction. The ferromagnetic signal often blinds us to the nature of the magnetic ground state. In contrast, in Ba$_2$IrO$_4$
such a ferromagnetic signal was not observed in all the temperature range (2 K–400 K). It is therefore considered that Ba$_2$IrO$_4$ is more suitable for studying the nature of SO Mott state than Sr$_2$IrO$_4$.

In this paper, we report on results of the muon spin rotation (μSR) study of Ba$_2$IrO$_4$. In general, muon is a sensitive microscopic probe to detect the internal local magnetic fields in solids. The μSR technique contributed to early progress in studies of high-$T_c$ cuprates, by determining the antiferromagnetic ground state of La$_2$CuO$_4$. We discuss the magnetic ground state and moment of the iridium ions in Ba$_2$IrO$_4$ based on the μSR spectra.

2. Experimental procedure
Polycrystalline samples of Ba$_2$IrO$_4$ were synthesized under 6 GPa of pressure using a flat-belt-type high-pressure apparatus installed at the National Institute for Materials Science (NIMS). The details of synthesis and characterization procedure were reported elsewhere[2]. Zero-field μSR experiments were performed using the D1 instrument at MUSE (Muon Science Establishment) in J-PARC, Tokai, Japan. Nearly 100% spin-polarized positive muons with a momentum of 28 MeV/c were implanted into the polycrystalline samples and the time-dependent muon spin polarization was measured using positron detectors. The samples were wrapped in a silver foil and mounted on a silver sample holder. Residual fields at the sample position were compensated within 1 μT in all directions by using correction coils and a flux-gate magnetometer.

3. Results
Figure 1 shows the zero-field μSR (ZF-μSR) time spectra measured at various temperatures. The positron asymmetry function $A(t)$ of the μSR signal rapidly decays near 240 K, when the temperature is lowered. This is an indication of the magnetic-order transition, where muons are susceptible to the paramagnetic random spins due to their critical slowdown. Below 240 K, muon-spin precession was observed. This is clear evidence that there exists a coherent internal magnetic field induced by an antiferromagnetic long-range order, because no spontaneous magnetization was observed in the previous magnetic susceptibility measurement [2]. The solid wavy curves indicate a numerical fit of the data using the exponentially damped oscillations with a slowly relaxing exponential function.

![Figure 1](image-url)

*Figure 1.* Selected zero-field μSR (ZF-μSR) time spectra measured at various temperatures in Ba$_2$IrO$_4$. The wavy curves indicate a numerical fit of the data using the exponentially damped oscillations with a slowly relaxing exponential function.

![Figure 2](image-url)

*Figure 2.* Temperature dependence of the precession frequency $f$. $B_\mu$ (right axis) denotes the local magnetic field. The solid line indicates the best fitting result (see text). The inset shows the real part of the fast Fourier transform (FFT) of the time spectra at 209 K.
Figure 2 shows the precession frequency \( f = \gamma B_\mu /2 \pi \), where \( \gamma = 2\pi \times 135.54 \, \text{MHz/T} \) is the muon gyromagnetic ratio and \( B_\mu \) is the local magnetic field at various temperatures. At 240 K, \( f \) is observed and increases rapidly, indicating that the antiferromagnetic long-range order transition occurs at \( T_N \sim 240 \, \text{K} \). The solid line indicates the best fitting result obtained by the phenomenological function \( f(T) = f(0) [1 - T/T_N]^{0.18} \) with \( f(0) = 2.67(6) \, \text{MHz} \) (corresponds to the internal local field \( B_\mu = 197 \, \text{G} \)), \( T_N = 243(1) \, \text{K} \), and \( \beta = 0.18(1) \). The inset of Fig.2 shows the real part of the fast Fourier transform (FFT) of the time spectra at 209 K. The single peak (~ 2 MHz) in the FFT result suggests that the implanted muons are mainly influenced by an internal field in unique \( \mu^+ \) site around \( T_N \).

4. Discussion

Implanted positive muons \( \mu^+ \) generally stop at the most electronegative site between atoms in solids, and interact with the internal local field \( B_\mu \) induced by the moment of the atoms at the \( \mu^+ \) site. In the case of oxides, \( \mu^+ \) usually stops near \( O^2- \) ions to form \((\text{O}^2^-\mu)^-\) bonds about 1 Å in length [3]. Figure 3 shows the schematic representation for possible \( \mu^+ \) site in \( \text{Ba}_2\text{IrO}_4 \). In \( \text{Ba}_2\text{IrO}_4 \), the \( \mu^+ \) site is presumed to locate at the most electronegative point with 1 Å apart from the apical oxygen site. As shown in Fig. 3, the \( \mu^+ \) site is specified by two numbers: the inclination angle \( \theta \) and the azimuth angle \( \phi \).

The electrostatic energy in (partially) ionic compounds is mainly composed of the Madelung site potential energy \( (E_M) \). We have calculated \( E_M \) on the spherical surface with radius 1 Å from the apical oxygen O2 (see Fig.3) to determine the most stable \( \mu^+ \) site. The Madelung site potentials have been calculated by using the Ewald summation method as implemented in the MADEL code [4]. The crystal structure parameters for \( \text{Ba}_2\text{IrO}_4 \) have been taken from the experimental work [2].

Figure 4(a) shows \( E_M \) versus \( \theta \) or \( \phi \) (see the inset of Fig.4(a)) plots at each \( \mu^+ \) site in \( \text{Ba}_2\text{IrO}_4 \). The calculated \( E_M \) gives the lowest potential at the site of \( \theta = 30^\circ \) and \( \phi = 0^\circ, 90^\circ \). The implanted \( \mu^+ \) are presumed to be placed around this site. The positive sign of \( \theta \) reflects a charge valence of anions between Ir\(^{4+}\) and Ba\(^{2+}\), in contrast with the negative \( \theta \) in \( \text{La}_2\text{CuO}_4 \) (Cu\(^{2+}\) and La\(^{3+}\)).

We calculated an effective magnetic moment of the iridium ions using a dipolar-field model, with an internal field of \( B_\mu = 197 \, \text{G} \). The dipolar field \( B_{\text{dip}} \) is given by

\[
B_{\text{dip}}(r) = \left(\mu_0/4\pi\right) \sum_i (3(\mathbf{\mu} \cdot \mathbf{n}_i)\mathbf{n}_i - \mathbf{\mu})/(r-r_i)^3,
\]

Figure 3. Schematic representation for possible \( \mu^+ \) site in \( \text{Ba}_2\text{IrO}_4 \). The \( \mu^+ \) site is specified by the inclination angle \( \theta \) and the azimuth angle \( \phi \).

Figure 4. (a) Madelung site potential energy (\( E_M \)) versus \( \theta \) or \( \phi \) (inset) plots. The minimum \( E_M \) is found at \( \theta = 30^\circ \) and \( \phi = 0^\circ, 90^\circ \). (b) \( \theta \)-dependence of the amplitude of the iridium moment \( |\mathbf{\mu}| \) with \( \phi \) fixed to \( 0^\circ, 90^\circ \).
where $\mu$ is the effective magnetic moment of the $i$-th Ir$^{4+}$ ion at $r_i$, $r$ is the $\mu^+$ site, $n_i = (r_i - r)/(|r_i - r|)$ is the unit vector from the $i$-th Ir$^{4+}$ ion to $\mu^+$, and $\mu_0$ is magnetic permeability of free space. For the calculation of the moment in Ba$_2$IrO$_4$, we assumed that the staggered moments in the antiferromagnetic ordered state are directed to the [110] or [-1-10] direction at the iridium sites. These assumptions are based on the findings of the systems La$_2$CuO$_4$ for the $\mu^+$ site [5] and of Sr$_2$IrO$_4$ for the direction of the iridium moments [6].

Figure 4(b) shows the $\theta$-dependence of the amplitude of the iridium moment $|\mu|$ at $T = 0$ K with $\phi$ fixed to 0°, 90°. At $\theta = 30^\circ$, $|\mu| = 0.34(4) \mu_B$/Ir-atom is obtained, where the number in the parenthesis indicates an error arising from an uncertainty of the $\mu^+$ site. Interestingly, the Ir$^{4+}$ moment (0.34 $\mu_B$) in Ba$_2$IrO$_4$ is much smaller than the integer moment $1 \mu_B$ expected in the case of $J_{\text{eff}} = 1/2$. This moment reduction is probably attributed to a low-dimensional quantum spin fluctuation with large intra-plane antiferromagnetic correlation $|J|$. A similar moment reduction is reported for parent materials of high-$T_C$ cuprate superconductors. In La$_2$CuO$_4$, the moment size of the Cu$^{2+}$ (3$d^9$, $S = 1/2$) ion is estimated to be $0.2 \sim 0.6 \mu_B$ from neutron scattering experiments [7, 8]. The reduced moment (0.34 $\mu_B$) in Ba$_2$IrO$_4$ is in good agreement with the values ($0.2 \sim 0.6 \mu_B$) in La$_2$CuO$_4$. The magnetic state in Ba$_2$IrO$_4$ is interesting in terms of an analogy to high-$T_C$ cuprate superconductors.

5. Summary
We measured magnetic properties of the spin-orbit Mott insulator Ba$_2$IrO$_4$ by muon spin rotation ($\mu$SR) experiments. Zero-field $\mu$SR experiments revealed that the magnetic ground state of Ba$_2$IrO$_4$ is antiferromagnetic long-range order ($T_N \sim 240$ K). In the antiferromagnetic state, the magnetic moment of the iridium ions is significantly reduced ($\sim 0.34 \mu_B$), indicating a low-dimensional quantum spin fluctuation with a large intra-plane correlation $|J|$. The magnetic state is quite similar to those in parent materials of high-$T_C$ cuprate superconductors such as La$_2$CuO$_4$.

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