DFT investigations on magnetic properties with Muon in La_2CuO_4 using LSDA + U functional

S. Charoenphon,[*1,*2] P. Reunchan,[*1] and I. Watanabe[*2]

Lanthanum cuprate (La_2CuO_4, LCO) is a typical Mott insulator exhibiting a strong covalent state of Cu-3d with O-2p within the two-dimensional (2D) CuO_2 plane. In a previous study, LCO has been well investigated experimentally and theoretically; however, questions regarding electromagnetic states remain unanswered. The primary concern is the on-site Coulomb potential, U, and the covalent state of the electronic orbitals of Cu. The muon spin resonance technique (μSR) can provide information on local magnetic fields at the muon site. However, obtaining the muon effect in the structure.[5)] In this study, we apply the density functional theory (DFT) + U to comprehensively investigate the electronic structure surrounding the muon. Our attempt to solve the problem within the DFT framework has yielded mixed results. Notably, most studies have attempted to correct the antiferromagnetic (AFM) ground state model of LCO and have failed to provide a handle on the key experimentally observed properties of LCO. More specifically, the local-spin-density approximation (LSDA) incorrectly predicts LCO to be nonmagnetic (NM) metals in complete disagreement with the experimental value. The generalized gradient approximation (GGA) produces a weak AFM. Thus, the development of methods that have stronger electron correlations to stabilize the AFM ground state is necessary. DFT + U is a method that was proposed to improve the strong correlation system.[6)] Among the most implemented methods in the DFT + U realm is the LSDA + U method.

The DFT calculation was performed using the RIKEN-integrated HOKUSAI Supercomputing Facility. Ab initio calculations were performed in the Vienna ab initio simulation package (VASP) with an energy cutoff of 400 eV for the plane-wave basis set. We employed the Hubbard parameter U = 4.87 eV.[4)] We use a 28-atoms primitive cell with a $6\times6\times3$ Γ-centered k-point mesh for the Brillouin zone integrations to calculate the fundamental properties of LCO shown in Fig. 1(a). In addition, a supercell structure containing 252 atoms ($3\times3\times1$ unit cell) was adopted for a calculation to consider the effect of the relaxation of muon position.

The lattice parameters calculated based on LSDA + U, band gap, and magnetic moment are listed in Table 1. The experimental values. The calculated bandgap was obtained as 1.15 eV, slightly higher than the experimental value of 1 eV.[3)] Next step, the LSDA + U was conducted, including the muon, to reproduce the μSR results. We estimate the initial position of muon injection in LCO obtained from the electrostatic potential. There are three possible local minimum potential positions that the muons can stop in a structure: M1, M2, and M3 as shown in Fig. 1(c), compared with muon spin resonance technique μSR results.[4,5)] Our method is fundamentally similar to the GGA functional method[4)] except for the difference in the functional. This is important for investigating the effect of the difference in the functional in the previous result.

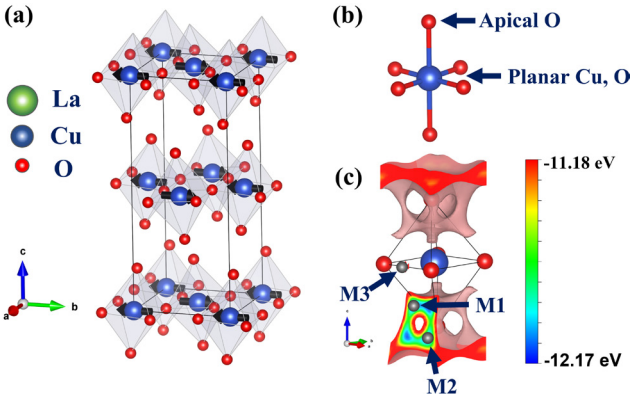

Fig. 1. (a) Crystal structure of LCO in the LTO phase with copper, oxygen, and lanthanum atom indicated as blue, red, and green spheres, respectively. (b) Schematic of a CuO_6 octahedral showing the O apical site and Cu, O in the planar site. (c) Electrostatic potential calculation result. The grey sphere indicates three local-minimum potential positions as candidates for initial muon-stopped positions.

Table 1. Calculation of structural parameter, band gap, and magnetic moment of a unit cell of LCO.

Function	Lattice parameter			The band gap (eV)	Magnetic moment (μ_B)
	a	b	c		
Exp.	5.357[1]	5.406[1]	13.143[1]	1[3]	0.48 ± 0.15[2]
LSDA+U	5.196	5.246	12.903	1.15	0.469

The final muon position and local deformations of the crystal structure estimated from our LSDA + U + μ calculations are shown in Fig. 2. We calculated the magnetic moment of Cu without muon to be 0.450 μ_B. Following relaxation, the magnetic moment calculation of Cu changed to 0.414, 0.453, and 0.449 μ_B for M1, M2, and M3, respectively.

*1 RIKEN Nishina Center
*2 Department of Physics, Kasetsart University

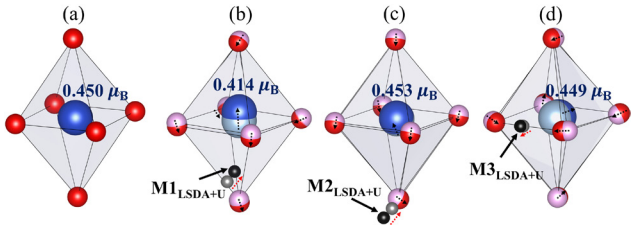

Fig. 2. (a) Structure of LCO without muon. The final muon position after relaxation for (b) M1, (c) M2, and (d) M3, respectively. The Grey spheres indicate the initial position. The black spheres indicate the final position of the muon after relaxation.

In conclusion, the role of muon perturbation was found to deform the local crystal structure in the immediate vicinity of the muon. This effect resulted in a slight reduction in the magnetic moment surrounding the muon. Moreover, a new result is that LSDA + U is clearly proven to be not suitable for describing the electronic state of a strongly correlated system, particularly LCO.

References

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