Quantum effects of muon on the electronic state of La_2CuO_4

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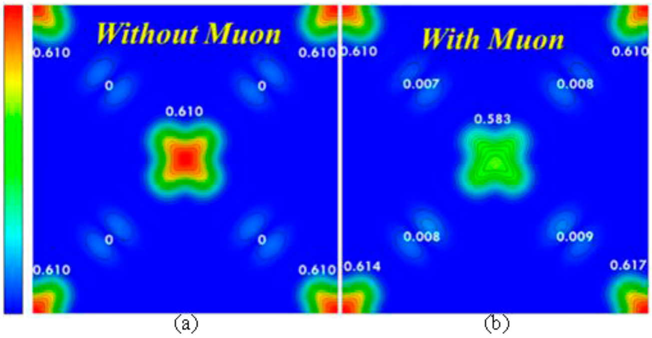
Muon spin relaxation (μSR) studies on La_2CuO_4 (LCO) prove the appearance of the antiferromagnetic (AF) long-range ordering as previously observed by powder neutron diffraction experiments.[1] The appearance of coherent muon-spin precession was observed and the internal field at the muon site was determined to be approximately 410 G.[2] Neutron diffraction experiments determined the AF spin structure, where the Cu spin had the magnetic moment of 0.5 μ_B aligning to the b-axis in the CuO_2 plane. Recent μSR studies on the thin film reported the observation of another muon-spin precession component, which experienced less internal field at the muon site of approximately 100–120 G.[3] Although both μSR and neutron diffraction experiments sense the same Cu spin in LCO, it is not yet possible to explain the internal fields at the muon site by using the AF spin structure as proposed by neutron diffraction experiments because the position of the implanted muon has not been precisely determined to explain the electronic state.

To deduce more detailed information and knowledge from the μSR results, we are developing techniques to more precisely estimate the muon position in LCO using the density functional theory (DFT) calculations. The Hubbard parameter U and exchange parameter J were set to be 8 and 0.8 eV, respectively. The AF spin structure proposed by Vaknin *et al.* was considered in our non-collinear calculations. To obtain a realistic behavior of muon, we utilized a large 32-unit supercell with a single muon implanted in the lowest position as shown by the electron potential map as reported in the previous study,[4] resulting in the perturbed system. We also consider the unperturbed system, which is the same supercell with no implanted muon to capture the essential differences between the perturbed and unperturbed systems. The ground state of the supercell was achieved by setting the convergence criterion to be 1×10^{-5} eV and relaxing all atoms until the magnitude of the force on each atom became less than 0.05 eV/Å. The dipole field value perceived by muon was evaluated using the spin-density grids acquired from our DFT calculations, given by the following equation:

$$H_{dip}(\vec{r}) = \sum_i \frac{1}{|\vec{r}-\vec{r}_i|}\left[3({\vec{\rho}_i}^{\,S}\cdot(\vec{r}-\vec{r}_i))\frac{(\vec{r}-\vec{r}_i)}{|\vec{r}-\vec{r}_i|^2}-{\vec{\rho}_i}^{\,S}\right] \qquad (1)$$

Where ${\vec{\rho}_i}^{\,S}$ is the magnetic moment of the Cu spin and $|\vec{r}-\vec{r}_i|$ is the relative distance between the muon and Cu spins. The total internal field at the muon site, $H_{dip}(\vec{r})$, is obtained from the summation of all dipole fields due to the surrounding Cu spins. For our calculations, we set the spherical region centered by the muon with a radius of 50 Å. This spherical region is sufficient for the convergence of the calculated internal fields.



(a) (b)

Fig. 1. Comparison of spin density map of LCO for (a) unperturbed system and (b) perturbed system.

Figure 1 shows the spin density contour map between the unperturbed (a) and perturbed (b) systems. Our calculations indicate that the unperturbed system is an AF insulator with 0.61 μ_B and an insulating gap $\sim$ 2 eV, which significantly corresponds with the experimental data.[5] The perturbed system, however, shows a local deformation in both crystal and spin structure owing the implanted muon. The nearest Cu spins are affected by the presence of the muon, thereby slightly reducing the value of Cu magnetic moment to 0.58 μ_B. These local deformations do not affect the electronic structure in general as the muon only affects the surrounding atom, making a large supercell necessary in our studies. Using Eq. (1), we evaluated the dipole field value of the muon inside our perturbed system and obtained a value of 498 G, which is close to the experimental value of 410 G. The difference of $\sim$ 80 G can be attributed to two possible issues. The first issue is the consideration of the zero-point vibrational motion for the implanted muon. The second is the utilization of a more optimized supercell structure to get a better description on how the implanted positive muons affect the crystal and spin structure. We believe that these two key points are mandatory to obtain more information from the muon's position inside the system.

References

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