Zero-point vibrational energy in the muon sites of La_2CuO_4

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La_2CuO_4 (LCO), the parent compound of high-T_C superconducting $La_{2-x}Ba/Sr_xCuO_4$, is known to have antiferromagnetic long-range ordering (AF-LRO) due to its classification as a Mott insulator. One accurate experiment to prove the AF-LRO characteristic is the muon spin relaxation (μSR) technique. In this technique, the implanted muons that sense an internal magnetic field undergo Larmor precession. A previous study on bulk LCO showed that the muon's precession frequency is 5.5 MHz. Using the muon's gyromagnetic ratio, $\gamma_\mu = 135.538817$ MHz/T, the internal field (B_μ^{exp}) felt by muons in bulk LCO was determined to be approximately 410 G.[1)] A recent study on thin-film LCO showed that the implanted muons probe an additional internal field approximately 100–120 G.[2)] While two clear precessions have been shown in the past, the exact location of the implanted muon is still unknown, which hinders the ability of the μSR technique to reveal more information and knowledge from the LCO system.

Our group has been developing a technique to precisely estimate the muon position by utilizing density functional theory (DFT) calculations. Generalized gradient approximation (GGA) was chosen for the exchange-correlation functional. The antiferromagnetic configuration of the Cu spin from Vaknin *et al.* is utilized for the non-collinear calculation.[3)] Because LCO is a Mott insulator, we need to include the Coulombic repulsion energy ($U_{eff} = 7.2$ eV) in the DFT calculation. From our previous reports,[4,5)] we show the muon position from our DFT calculation and the muon's perturbation effect on the Cu-spin densities from a DFT perspective. However, even when using the distributed-spin model to calculate the internal field, the calculated internal field shows a 21% difference with that observed from the experimentally ($B_\mu^{exp} = 410$ G, while $B_\mu^{DFT} = 498$ G). We considered that this discrepancy originates from the zero-point vibrational motion (ZPVM) of the muon, which was not considered in our last report. As a fine quantum particle, the muon should fluctuate in its lowest-energy state. To include the ZPVM, we first evaluate the three-dimensional Hartree potential ($V_\mu^{Hartree}$) around the implanted muon's position. Subsequently, we simply calculate the eigenvalues and eigenfunction for the ground state on those areas by using the following equation:

$$\left[\frac{\hbar^2}{2m_\mu}\nabla_i^2 + V_\mu^{Hartree}(r)\right]\Psi_\mu(r) = E_\mu \Psi_\mu(r), \qquad (1)$$

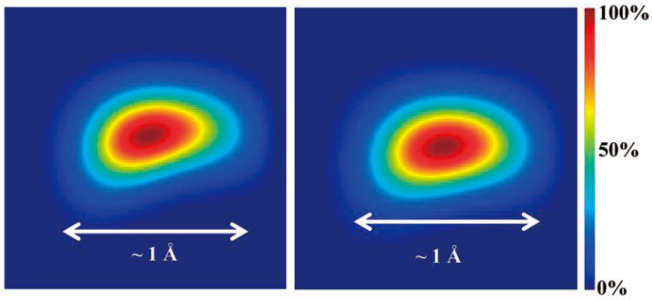

Fig. 1. Muon probability density map in the xz-plane for the (a) unperturbed LCO (b) perturbed LCO.

where m_μ denotes to the muon mass, and $|\Psi_\mu|^2(r)$ is the probability that the muon exists at each position r. This Schrödinger equation was using the finite-difference method. From our calculations, we conclude that the energy convergence is achieved by considering a $1.5 \times 1.5 \times 1.5$ (Å) area from the center of the muon position. The probability that the muon exists within this designated area is 100%.

As reported previously, the implanted muon only affects the local crystal and electronic structure of LCO. From our calculations, we obtained the ground-state energy as 0.82 eV for the unperturbed LCO and 0.89 eV for the perturbed LCO. The slight increase of the ground-state energy is very likely due to the local deformation of the crystal structure. However, the general description of the muon's distribution over the considered area is relatively the same as shown in Fig. 1. By using the muon probability density map of the perturbed LCO and the distributed-spin model from our previous reports[4,5)], we successfully reduced the differences of the internal field to less than 1% ($B_\mu^{exp} = 410$ G, while $B_\mu^{DFT+ZPVM} = 409$ G). While this method shows a promising result for the LCO system, it still needs to be tested in other systems, in order to standardize this method to assist in μSR experiments.

References
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