Estimation of the on-site Coulomb potential in La_2CuO_4: A μSR and DFT Study

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Since its discovery in the late 1980s, high-temperature superconducting cuprates are known to have a rich variety of physics and have attracted interest from researchers worldwide. There are open questions on exotic electronic states that need to be investigated, such as pseudogaps, stripes of spins and holes, and charge-ordered states. These properties are usually described on the basis of the strong on-site Coulomb potential, U, and the covalent states of the Cu 3d orbitals with the surrounding O 2p orbitals. To understand more about these properties, La_2CuO_4 (LCO) can be used as the ideal candidate material. LCO is the parent compound of high-Tc superconducting is classified as a Mott insulator, and has strong covalent states between the Cu 3d and O 2p orbitals. These interactions produce antiferromagnetic insulating behavior due to the existence of the on-site Coulomb potential energy (U). The value of U of this system has been thoroughly investigated but still has large ambiguities in the range of 3–10 eV, raising uncertainties in the discussion of the electronic states in high-Tc superconducting cuprates.

Our group has been developing a technique to estimate the value of U by combining the muon spin rotation (μSR) and density functional theory (DFT) and including U as an adjustable parameter (DFT + U). From an experimental perspective, a muon is a sensitive local magnetic probe that can be used to observe the ordered state of LCO. Our group has shown that an implanted muon has three frequency components using the high-statistic μSR ,as shown in Fig. 1. These three frequency components can be translated into three internal fields of 426.7(1), 109.2(4), and 1251.6(3) G.[1] From a theoretical perspective, we develop a method to obtain a more precise muon position by utilizing DFT + U. From calculations, we show that there are three possible positions at which a muon can reside. As discussed in the previous APR report, we expand the calculation further by considering the local effect of an implanted muon from both the crystal and spin structures of supercell LCO.[2,3] After that, we also consider the zero-point vibration motion (ZPVM) of an implanted muon inside the structure by calculating the distribution of the muon position as a quantum particle.[4] Both of these considerations can be utilized to calculate the internal field by using the following equation:

$$\sum_{i,j} \frac{1}{|\vec{r}_i-\vec{r}_j|^3}\left[3\vec{\rho}_i\left(\vec{r}_i-\vec{r}_j\right)\frac{\left(\vec{r}_i-\vec{r}_j\right)}{|\vec{r}_i-\vec{r}_j|^2}-\vec{\rho}\right]|\psi_j|^2 . \quad (1)$$

Here, $\vec{\rho}_i$ denotes the vector data for the spin grids and

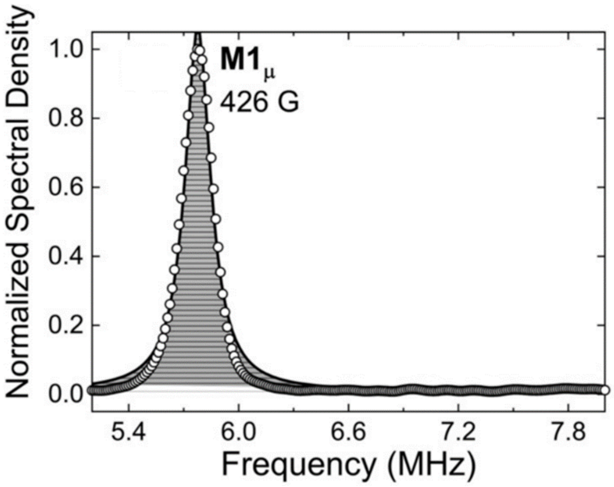

Fig. 1. Fourier transfrom spectrum from the high-statistic μSR experiment.

$\vec{r}_i - \vec{r}_j$ denotes the relative distance between Cu-spin grids with a density of $\vec{\rho}_i$ and the muon distribution grids ψ_j. Then, we sum all grid components obtained from our DFT calculations to estimate the internal field at each muon, a sphere with a radius of 50 Å. After obtaining the internal fields for each fields muon position, we optimize U by comparing the calculated internal fields with the experimental internal fields:

$$\varDelta H_{M_i} = \left(H^{M_i}_{DFT} - H^{M_i}_{\mu SR}\right), \ |i = 1, 2, 3|, \quad (2)$$

and sum all $\varDelta H_{M_i}$ for each U with the fitting errors of the internal fields, σ_i, as follows:

$$\sum_i \frac{\varDelta H^2_{M_i}}{\sigma_i^2}, \quad |i = 1, 2, 3|. \quad (3)$$

By applying the Gaussian function, we obtain optimized U of 4.87(4) eV. By using this optimized U, we observe that the difference between the calculated and experimental internal fields for the M1, M2, and M3 positions are 3.4 (≈1%) , 38.2 (≈40%), and 97.9 (≈8%) G respectively.[1]

References
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