Positron potential and wavefunction in LaFeAsO

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We report calculations of the positron potential and wavefunction in LaFeAsO. These calculations show that the positron wavefunction does sample the entire unit cell although it is largest in the interstices of the La layer adjacent to As atoms. The implication is that angular correlation of annihilation radiation (ACAR) is a viable probe of the Fermi surfaces in this material. The results also apply to positive muons, and indicate that these will be localized in the La layer adjacent to As.

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I. INTRODUCTION

The discovery of high temperature superconductivity in a family of layered oxypnictides, prototype LaFeAs(O,F) with critical temperatures exceeded only by cuprates has stimulated considerable interest both in determining the chemical dependence of the properties and in understanding the mechanism for superconductivity. Central to this discussion is the underlying electronic structure.

These materials occur in a tetragonal structure based on a square lattice of Fe coordinated in such a way that the unit cell is based on a (2x2) doubling of the Fe square lattice structure (see Fig. 1). First principles calculations done within density functional theory predict five small sheets of Fermi surface in the undoped compound, LaFeAsO: two 2D electron cylinders at the zone corner (M), two heavier 2D hole cylinders at the zone center (Γ) and a still heavier 3D hole pocket, which intersects the hole cylinders. Because of the heavy masses of the bands, especially the hole bands, the density of states is high even though the carrier density is low.

Superconductivity generally requires a pairing interaction. Direct calculations of the electron-phonon interaction have shown that it is modest (the coupling is λ ~0.2) and in particular is far too weak to explain the observed critical temperatures. In unconventional superconductors the k dependence of the pairing interaction plays a central role as this k dependence acting on the Fermi surface determines the symmetry of the superconducting state that emerges. The 2D cylinders are roughly nested and as such may be expected to lead to k dependence of properties. In fact a spin density wave associated with the nesting has been predicted and observed in the undoped compound but not so far in the doped superconducting material. Models of superconductivity associated with spin fluctuations deriving from this nesting have been discussed. However, small Fermi surfaces, which are derived from states near band edges, are in general particularly sensitive to details of the crystal structure and are also more sensitive to disorder and perhaps to many body effects. As such, it is important to determine the Fermi surface from experiment.

The most common experimental probes of Fermi surfaces in metals are (1) quantum oscillation measurements, such as de Haas van Alphen and Shubnikov de Haas, (2) angle resolved photoelectron spectroscopies (ARPES), (3) positron annihilation, in particular angular correlation of positron annihilation radiation (ACAR) and (4) Compton scattering. These techniques are complimentary. Quantum oscillation techniques are the methods of choice since they are direct and have the highest resolution when they are practical, but they require very high quality samples with long mean free paths. ARPES is particularly applicable to 2D materials, where it gives a direct map of the Fermi surface and does not require such high mean free paths. It is however surface sensitive and requires that clean unreconstructed surfaces characteristic of bulk can be made. Positron ACAR and Compton scattering are bulk techniques with lower resolution than ARPES or quantum oscillations, but with much less stringent sample quality issues. ACAR is the more common of these two techniques and may be the most readily applicable method if crystals of the LaFeAs(O,F) phases become available. However, the sensitivity of ACAR measurements depends
FIG. 2: (Color online) Inverted Coulomb potential of LaFeAsO shown in (110) planes (see Fig. 1). The potential is divergently repulsive inside the atom cores. The contours shown span a range of 5.65 eV and are equally spaced.

FIG. 3: (Color online) Positron potential of LaFeAsO (see Fig. 1). The potential is divergently repulsive inside the atom cores. The contours span a range of 5.92 eV and are equally spaced.

on the overlap of the positron wavefunction with the electronic states at the Fermi level. In these materials those states are primarily Fe d states modestly hybridized with As p states. A related issue was recognized in high $T_c$ cuprate superconductors, where it was found that the sensitivity of ACAR to CuO$_2$ planes depended on the particular material, and in particular that ACAR was not sensitive to the CuO$_2$ derived electronic states in YBa$_2$Cu$_3$O$_7$. The purpose of the present paper is to report positron wavefunctions for LaFeAsO.

II. APPROACH

The present calculations were done using the general potential linearized augmented planewave method including local orbitals. The self consistent electronic structure was first calculated using the experimental lattice parameters but the LDA relaxed internal coordinates for La and As. Computational parameters for this were set as described in Ref. 4. The positron calculation was then done using the potential generated from the electronic charge density. In particular, the bulk positron wavefunction was computed in the inverted Coulomb potential to which a correlation term was added,

$$V^+|n| = -V_{\text{Coul}}|n| + V_{\text{corr}}|n|,$$

where $V^+$ is the potential to be used for the positron wavefunction calculations, $V_{\text{Coul}}$ is the Coulomb potential from the electronic calculation and $V_{\text{corr}}$ is an electron positron correlation function. All of these are functionals of the electron density, but not the positron density, since for the bulk positron wavefunction a single positron is distributed over a macroscopic sample, and therefore has vanishing density. The correlation function, $V_{\text{corr}}$ is an attractive potential that arises from the response of the electrons to the positron. While the sum rule that the correlation peak, which is the analogy of the exchange-correlation hole of the electron gas, has unit charge, the magnitude of this buildup is not bounded, while the electron-electron hole is bounded by the local density. Therefore, the electron-positron correlation potential is thought to be less well behaved than the electron-electron correlation. Nonetheless, local density parameterizations have been developed. Here we use the parameterization of Boronski and Nieminen,14 applied as in Ref. 15. We note that a similar approach can in principle be applied to other charged particles, in particular positive muons, $\mu^+$. In that case the potential for the $\mu^+$ can be formally written as for the positron, except that because of the higher mass of the $\mu^+$ the reduced mass of an $\mu^+$ - e$^-$ pair is higher, and therefore the correlation potential will be stronger and more difficult to reliably parameterize.16,17,18

III. RESULTS AND DISCUSSION

The main results of this work are shown in Figs. 2, 3 and 4. These show respectively the inverted Coulomb potential, the positron potential, $V^+$ and the positron density, which is the square of the positron wavefunction, normalized to one positron per cell for convenience.

As may be seen, the inverted Coulomb potential is most attractive in the interstices of the Fe-As part of the unit cell. A positron localized there would be most sensitive to the electronic states near the Fermi energy. However, the electron density is higher on the other side of the As ions, in the interstices of the the La layer. Because of this, the addition of the correlation term favors
FIG. 4: (Color online) Positron density of LaFeAsO (see Fig. 1). The positron density goes to zero in the atom cores. The contours are equally spaced and range from 0.00008 to 0.00265 in units of positrons per cubic Bohr, normalized to one positron per unit cell.

that site. The absolute minimum of the potential remains near the Fe when the correlation potential is added. However, the position in the interstices of the La layer above As is larger, and the additional attraction provided by the correlation function is enough to shift the maximum of the positron density to that position. Still, this is not strong enough to fully pull the positron density away from the Fe planes. Thus as may be seen in Fig. 4 the positron density is highest in the interstices of the La plane nearest to As following the potential, \( V^+ \). The calculated positron lifetime, obtained as in Ref. 15, is 163 ps.

This result means that positrons will mainly sample the electronic states away from Fe. However, this is not as strong a localization as was found in YBa\(_2\)Cu\(_3\)O\(_7\) (Ref. 11), and in particular some significant density may still be seen around the Fe. Furthermore, it should be noted that the states near the Fermi level have some hybridization between Fe and As, similar to an oxide electronic structure. As such, the fact that the positrons are sensitive to As will give them some sensitivity to the Fermi surface. Thus is would seem that ACAR is a viable technique for detecting the Fermi surfaces of LaFeAs(O,F).

Turning to positive muons, there have been several recent muon spin rotation studies of these materials.\(^{19,20,21}\) These studies have yielded quite useful insights into the magnetism of the materials, showing signatures both of the spin density wave and of rare earth magnetism and in addition have been useful in establishing the penetration depth. As mentioned the correlation potential for muons will be more strongly attractive than for positrons. Furthermore, because of their heavier mass, muons will not be delocalized in the lattice, but rather will localize, similar to a proton. This will lead to a change in the electron density, which should be treated self-consistently. Qualitatively however this effect will also amount to an increased tendency for the muon to be located where the electron density is high. This means that the effect that for the positron adding \( V_{\text{corr}} \) to the inverted Coulomb potential draws the positron away from the Fe layer will be enhanced for positive muons so that they will be drawn away even more strongly. Thus we may conclude that positive muons probe the interstices in the La layer adjacent to As, and therefore that are most sensitive to the rare earth site.

IV. SUMMARY

Density functional calculations of the positron wave-function in LaFeAsO show that positrons probe the entire unit cell, but are mainly located in the interstices of the La layer adjacent to As in this structure. Considering that there is some overlap with the Fe layer and considering that the Fe \( d \) derived electronic states at the Fermi energy are hybridized with As, albeit modestly, we conclude that positron ACAR can be used to measure Fermi surfaces in this material.

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