Positron potential and wave function in LaFeAsO

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We report calculations of the positron potential and wave function in LaFeAsO. These calculations show that the positron wave function 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 annihilation correlation of annihilation radiation 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[1] of high-temperature superconductivity in a family of layered oxypnictides, prototype LaFeAs(O,F) with critical temperatures exceeded only by those of 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,[1–3] based on a square lattice of Fe coordinated in such a way that the unit cell is based on a $c(2\times2)$ 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 two-dimensional (2D) electron cylinders at the zone corner (M), two heavier 2D hole cylinders at the zone center (Γ), and a still heavier three-dimensional (3D) hole pocket, which intersects the hole cylinders.[4] 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 $\lambda\sim0.2$) and in particular is far too weak to explain the observed critical temperatures.[5,6] 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.[6–9] Models of superconductivity associated with spin fluctuations deriving from this nesting have been discussed.[6] However, small Fermi surfaces, which are derived from states near band edges, are in general particularly sensitive to the 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 spectroscopy (ARPES); (3) positron annihilation, in particular angular correlation of positron annihilation radiation (ACAR); and (4) Compton scattering. These techniques are complementary. Quantum oscillation techniques are the methods of choice since they are direct and have the highest resolution when they are practical. However 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 that of ARPES or quantum oscillations but with much less stringent sample quality issues, although it should be noted that positrons are often sensitive to vacancy type defects. ACAR is the more common of these two techniques and may be the most readily applicable method when crystals of the LaFeAs(O,F) phases are available. However, the sensitivity of ACAR measurements depends on the overlap of the positron wave function with the electronic states at the Fermi level. In these materials those states are primarily Fe d states modestly hybridized with As p states. The corresponding issue was recognized in high-T_c cuprate superconductors.[10–14] Specifically, for a Fermi surface to be detected by ACAR, there must be suffi-

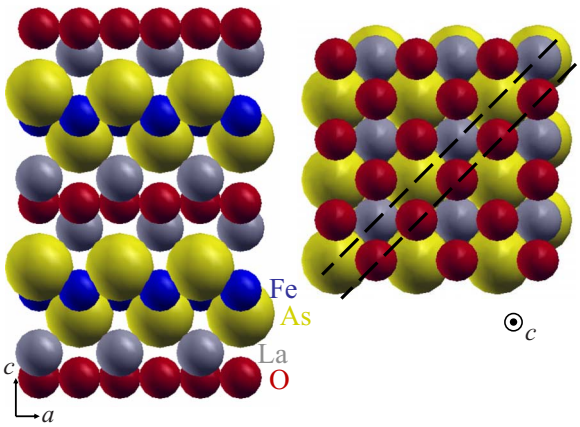

FIG. 1. (Color online) Structure of LaFeAsO viewed along [100] (left) and [001] (right) directions. The dashed lines in the right panel show the two cuts for which potentials and positron densities are plotted in Figs. 2–4.

cient overlap between the positron wave function and the electronic states associated with the band crossing the Fermi level. In simple materials, such as elemental metals and many intermetallics, this is the case as the positron wave function samples the entire interstitial region of the unit cell. However, in complex materials this need not be the case, and the positron may be confined to one region of the unit cell, while the electronic states of interest may be associated with atoms in another part of the cell. In the cuprates, the Fermi surfaces and electronic states of most interest are those associated with the CuO_2 planes. However, it was found that in $YBa_2Cu_3O_7$ the positron wave function is confined to the so-called chain region of the unit cell and has very little overlap with the CuO_2 planes. On the other hand in other materials, particularly the Tl series and the so-called 214 compounds (e.g., $La_{2-x}Sr_xCuO_4$),[11,13,14] there is overlap between the positron wave function and the CuO_2 planes, which both allows positrons to detect the relevant electronic states and also provides sensitivity to other features of the planes, in particular magnetism.

The purpose of the present Brief Report is to report positron wave functions for LaFeAsO. We find that there is indeed overlap between the positron wave function and the Fe-As part of the unit cell and therefore that it should be possible to detect the Fermi surfaces using ACAR. ACAR should also be sensitive to changes in the Fermi surface due to magnetism, which is thought to be an important part of the physics of the Fe-As materials.

II. APPROACH

The present calculations were done using the general potential linearized augmented plane-wave method including local orbitals.[15,16] The self-consistent electronic structure was first calculated using the experimental lattice parameters with the local-density approximation (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 wave function was computed in the inverted Coulomb potential, to which a correlation term was added,

$$V^+[n] = -V_C[n] + V_c[n], \tag{1}$$

where V^+ is the potential to be used for the positron wave function calculations, V_C is the Coulomb potential from the electronic calculation, and V_c 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 wave function a single positron is distributed over a macroscopic sample and therefore has vanishing density. The correlation function V_c is an attractive potential that arises from the response of the electrons to the positron. Although the sum rule states 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 parametrizations have been developed. Here we use the parametrization of Boronski and Nieminen,[17] applied as in Ref. 18. We note that a similar approach can in principle be applied to other charged particles, in particular positive muons μ^+. In that case the potential for the μ^+ can be formally written as for the positron except that because of the higher mass of the μ^+, the reduced mass of an μ^+-e^- pair is higher. Therefore the correlation potential will be stronger and more difficult to reliably parametrize.[19–21]

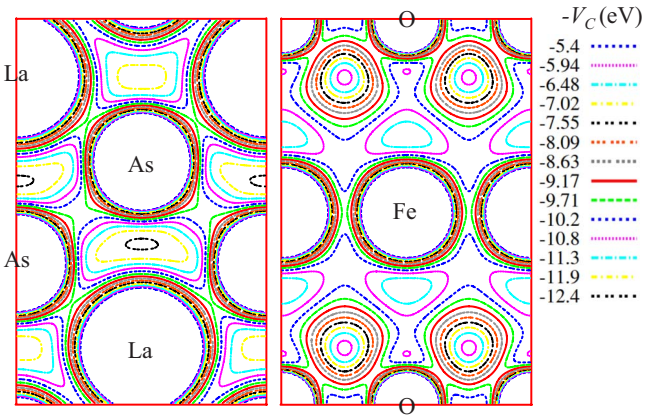

FIG. 2. (Color online) Inverted Coulomb potential $-V_C$ of LaFeAsO shown in (110) planes (see Fig. 1). The potential is divergently repulsive inside the atom cores. The contours shown span a range of 7 eV and are equally spaced. Note that the energy zero is arbitrary.

III. RESULTS AND DISCUSSION

The main results of this work are shown in Figs. 2–4. These show respectively the inverted Coulomb potential, the positron potential V^+, and the positron density, which is the square of the positron wave function, 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,

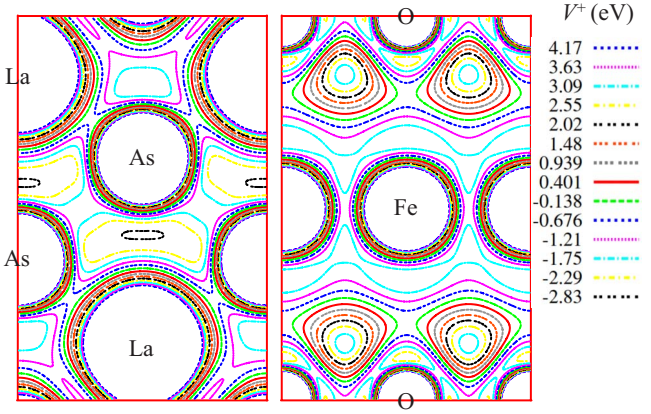

FIG. 3. (Color online) Positron potential V_c of LaFeAsO (see Fig. 1). The potential is divergently repulsive inside the atom cores. The contours span a range of 7 eV and are equally spaced. Note that the energy zero is arbitrary.

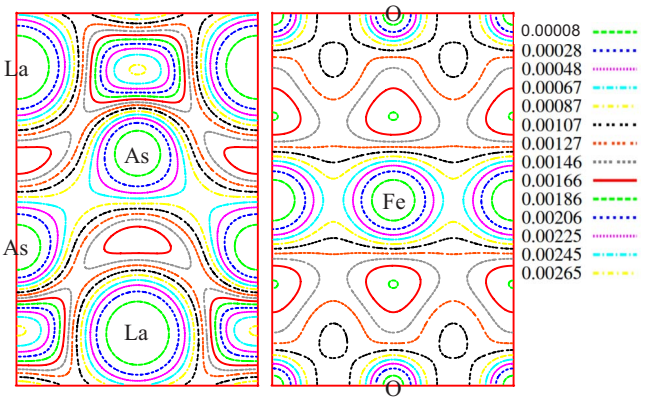

FIG. 4. (Color online) Positron density of LaFeAsO (see Fig. 1). The positron density goes to zero in the atom cores. The contours are equally spaced and range from 0.000 08 to 0.002 65 in units of positrons per cubic bohr, normalized to one positron per unit cell.

although there is an interstitial region in the La layer opposite As that is nearly as attractive. A positron localized in the Fe-As layer 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 La layer. Because of this, the most attractive part of the positron potential is shifted away from the Fe-As region, to a position in the interstices between As and La. However, this is not strong enough to fully pull the positron density away from the Fe planes. 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. 18, is 163 ps.

This result means that although positrons will mainly sample the electronic states away from Fe, they have substantial overlap with the Fe-As layers, in contrast to $YBa_2Cu_3O_7$ (Ref. 11). In particular some significant density is 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. 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.[22–24] 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_c 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 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. However, there is overlap with the Fe layer and additionally the Fe *d* derived electronic states at the Fermi energy are hybridized with As, albeit modestly. As such we conclude that positron ACAR can be used to measure Fermi surfaces in this material.

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