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# Detecting sign-changing superconducting gap in LiFeAs using quasiparticle interference

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Using a realistic ten-orbital tight-binding model Hamiltonian fitted to the angle-resolved photoemission (ARPES) data on LiFeAs, we analyze the temperature, frequency, and momentum dependencies of quasiparticle interference (QPI) to identify gap sign changes in a qualitative way, following our original proposal [Phys. Rev. B 92, 184513 (2015)]. We show that all features present for the simple two-band model for the sign-changing  $s_{+-}$ -wave superconducting gap employed previously are still present in the realistic tight-binding approximation and gap values observed experimentally. We discuss various superconducting gap structures proposed for LiFeAs, and identify various features of these superconducting gaps functions in the quasiparticle interference patterns. On the other hand, we show that it will be difficult to identify the more complicated possible sign structures of the hole pocket gaps in LiFeAs, due to the smallness of the pockets and the near proximity of two of the gap energies.

## I. INTRODUCTION

In the iron-based superconductors, the origin of the Cooper-pairing and the overall phase structure of the superconducting order parameter is still under debate. Nevertheless, taking into account the Fermi surface topology of iron based superconductors, there is a general consensus that the superconducting state in most of these materials belongs to the  $A_{1g}$  symmetry representation as long as both the electron and the hole Fermi surface pockets are present in the system<sup>1,2</sup>. At the same time, due to the multiplicity of the Fermi surface sheets and multi-orbital character of the electron states near the Fermi level, the actual phase structure of the superconducting gap distributed between various Fermi surface and orbitals is less clear. While orbital fluctuations together with the electron-phonon interaction favor the conventional “ $s_{++}$ ” wave superconducting gap without internal phase shifts<sup>3</sup>, spin fluctuations and enhanced interband repulsion originating from the proximity to the antiferromagnetic phase favor the so-called  $s_{+-}$ -wave structure in which the sign of the order parameter changes between the hole and electron bands.<sup>4-6</sup> Furthermore, the situation can be even richer if the magnetic fluctuations are not so strong as it is the case in LiFeAs<sup>7-9</sup>, or when fluctuations at more than one wavevector compete, as in FeSe<sup>10,11</sup>. In particular, it was argued that in FeSe the electronic renormalization effects for different orbitals result in orbital selectivity for the Cooper-pairing<sup>11,12</sup>, which in turn also affects the overall magnitude and the phase structure of the superconducting gap. In LiFeAs, orbital selective renormalization was also recently observed<sup>12,13</sup>. Flavours of the  $s_{+-}$  pairing symmetry where the phase of the order parameter is additionally changing between different hole or electron Fermi pockets have been proposed for this or other Fe-based superconductors<sup>8,9,14-16</sup>.

One rapidly developing technique to determine the phase structure of the order parameter makes use of quasiparticle interference as measured by Fourier transform scanning tunneling microscopy (FT-STM). This probe measures the wavelengths of Friedel oscillations caused by disorder present in a metallic or superconducting system, which in turn contains in-

formation on the electronic structure of the pure system. The subset of the scattering wave vectors  $\mathbf{q}$  can be enhanced or not according to the type of disorder and the phase structure of the superconducting gap, as noted by several groups<sup>17,18</sup> and experimentally verified in the cuprate superconductor,  $\text{Ca}_{2-x}\text{Na}_x\text{CuO}_2\text{Cl}_2$ <sup>19,20</sup>, where it was shown that QPI intensity at certain  $\mathbf{q}$  was selectively enhanced or suppressed by the external magnetic field, consistent with  $d_{x^2-y^2}$ -wave pairing. Later the same experiment was also performed for the  $\text{FeSe}_{0.4}\text{Te}_{0.6}$ <sup>21</sup>, although the interpretation of the results was less obvious due to the absence of observable vortices in this particular experiment.<sup>21-25</sup> This methodology is very efficient if an isolated set of features can be separated and monitored, as in the nodal  $d$ -wave symmetry. However, in multiband systems with complicated momentum dependence of the order parameter, it boils down to comparing multiple theoretically-generated complex patterns with the, usually noisy, experimental picture.

Recently, we proposed a novel method of analyzing the QPI data from non-magnetic impurities, which does not rely on the momentum dependence of individual features, but rather upon integrated indicators, averaged, formally over the entire momentum space. This method utilizes valuable phase information, discarded in usual treatments (see, however, Refs. 26 and 27). It was shown that some integrated indicators, specifically the antisymmetrized combination of the conductance, behave qualitatively differently for scattering events accompanied or not by a change in the order parameter sign. This method does not require manipulating scattering by magnetic field<sup>24</sup>, whose role as creator of artificial vortex “disorder” is unclear. The derivation in Ref. 24 does not call for any assumption about the structure of the order parameter in the momentum space, as long as it either has or does not have regions with the opposite gap signs. While this derivation was completely general, it was also confirmed by extensive numerical simulations with finite disorder<sup>28</sup>. Using this method, the authors of Ref. 11 were able to identify a sign changing order parameter in FeSe, and similar conclusions were recently drawn for Li hydroxide intercalated FeSe as well<sup>29</sup>.

In this connection, a question arises whether a more detailed analysis of the same characteristic, namely the antisymmetrized conductance, can be used to extract more information than just an overall sign change. LiFeAs, for which interesting possibilities, such as sign-flipping between the hole Fermi surfaces, have been suggested<sup>9</sup>, is therefore a natural candidate. With this goal in mind, we have extended our previous analysis to a realistic multi-orbital model of LiFeAs. The electronic structure of LiFeAs was previously measured by ARPES and fitted to a 10 orbital tight-binding Hamiltonian<sup>7</sup>. This provides rather comprehensive information on the orbital and band structure of this material in the low-energy regime. Furthermore, the superconducting gap values for each of the Fermi surface sheets were also measured both by ARPES<sup>30,31</sup> and previous STM measurements<sup>32-35</sup>. This provides an ideal testing ground for our earlier proposal given the high-quality STM data available for LiFeAs<sup>36-38</sup>. Note that for this material, the problem of the sign change of the gap between electron and hole pockets was studied earlier by Chi et al.<sup>32</sup> using ad hoc expressions for the LDOS based on BCS coherence factors, which were shown in Ref. 24 to be incorrect; nevertheless similar conclusions to [24] were reached. In addition, an attempt was made to provide a quantitative calculation of the STM spectra for various impurities such as Mn, Ni or Co and different gap symmetries<sup>39</sup>. In this manuscript, we present a qualitative analysis for realistic band structure, using the correct observable sensitive to gap sign change, but independent of the details of impurity wave functions.

As expected, we find that despite the complications introduced by the multiple Fermi surfaces, the distinction between the  $s_{++}$  and  $s_{+-}$  can be readily made based on the integrated antisymmetrized conductance. We further investigate how various symmetries, discussed in the literature, can be identified via characteristic  $\mathbf{q}$ -behavior of the antisymmetrized conductance and could be used for a finer elucidation of the gap sign structure in iron based materials up to detailed identification of the gap sign corresponding to different pockets. Specifically, it is tempting to separate electron-hole and hole-hole scattering features, and investigate the latter for potential signature of the sign change. As we show below, while such a separation is possible, proximity of the two gaps in the two main hole bands precludes a firm conclusion about possible sign change between them.

## II. NORMAL STATE BAND STRUCTURE AND QPI

We employ the ten-orbital tight-binding model Hamiltonian fitted to the ARPES measured band structure of LiFeAs samples by Wang et al.<sup>7</sup>, where each 5 orbitals correspond to one of the two Fe-atoms within the unit cell. The doping level of the system corresponds to  $n = 6 e^-/\text{Fe}$  and the resulting Fermi surface is shown in Fig. 1(a) and (b) for two values of  $k_z = 0$  and  $k_z = \pi$ . Due to the smallness of the Fermi energies for the small hole pockets of the  $xz, yz$ -character they appear to have stronger  $k_z$  dependence and close around the Z point of the Brillouin zone.

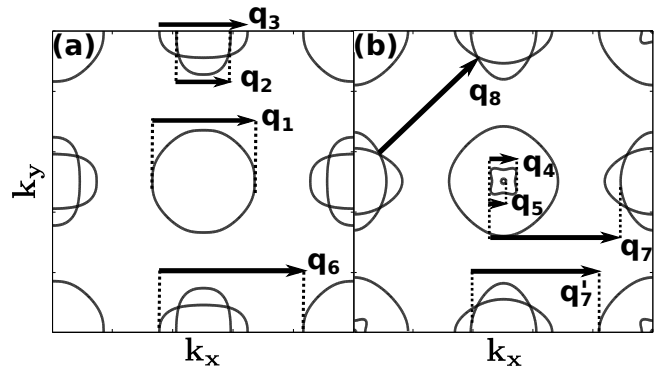


FIG. 1. Cuts of constant energy in LiFeAs for (a)  $k_z = 0$ , and (b)  $k_z = \pi$ , plotted in the 1-Fe Brillouin zone. Some potential scattering vectors  $\mathbf{q}_i$  are indicated by the arrows. Chemical potential chosen to (a)  $\mu = 3$  meV and (b)  $\mu = 1$  meV corresponding to  $n = 6$  electrons in the system.

The bare non-interacting Green's function is given by

$$\hat{G}^0(\mathbf{k}, \omega) = (i\omega_n - \hat{H}_0)^{-1}. \quad (1)$$

where  $\omega_n$  refers to the Matsubara frequency. All quantities may be considered to be matrices in band or orbital space, but the unperturbed Green's function is diagonal in band space. The local density of state (LDOS) is then found as  $\rho(\omega) = -\frac{1}{\pi} \text{Tr} \text{Im} \sum_{\mathbf{k}} \hat{G}^0(\mathbf{k}, \omega)$  where the Tr runs over the orbital or band index. In the presence of impurities the equation determining the full Green's function reads

$$\hat{G}(\mathbf{k}, \mathbf{k}', \omega) = \hat{G}^0(\mathbf{k}, \omega) + \hat{G}^0(\mathbf{k}, \omega) \hat{t}(\mathbf{k}, \mathbf{k}', \omega) \hat{G}^0(\mathbf{k}', \omega), \quad (2)$$

where the last term describes multiple scattering on impurities. In the following we consider multiple scattering from a single pointlike spherically symmetric non-magnetic impurity<sup>40,41</sup>, i.e. the impurity potential  $\hat{U}$  is taken to be well-localized and independent of spin. The  $t$ -matrix is then no longer momentum dependent, and is related to the impurity potential by  $\hat{t} = \hat{U} + \hat{U} \hat{G}^0 \hat{t}$ .

In such a case the solution for the  $t$ -matrix may be written,

$$\hat{t}(\omega) = [1 - \hat{U} \sum_{\mathbf{k}} \hat{G}^0(\mathbf{k}, \omega)]^{-1} \hat{U}. \quad (3)$$

Thus the position dependent correction to the LDOS reads may be written  $\rho(\mathbf{q}, \omega) = \rho(\omega) + \delta\rho(\mathbf{q}, \omega)$  with

$$\delta\rho(\mathbf{q}, \omega) = -\frac{1}{\pi} \text{Tr} \text{Im} \sum_{\mathbf{k}} \hat{G}^0(\mathbf{k}, \omega) \hat{t}(\omega) \hat{G}^0(\mathbf{k} + \mathbf{q}, \omega) \quad (4)$$

where in the last equation we employ the analytic continuation to the advanced (retarded) Green's functions.

We have not yet specified the form of the impurity potential in band or orbital space. Intuition suggests and density functional theory calculations confirm to a very good approximation that  $\hat{U}$  is diagonal in orbital space<sup>42</sup>. In the following, until specified otherwise we assume the impurity potential to be diagonal in the orbital space with equal amplitudes

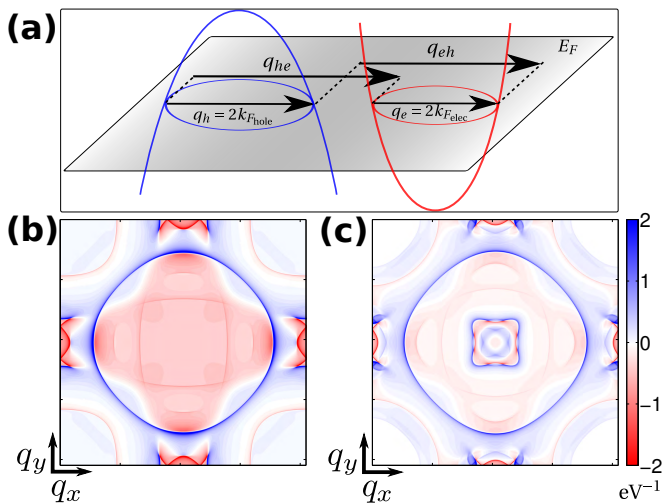


FIG. 2. (a) Schematic representation of possible scattering events in a one electron and one hole pocket system. (b),(c) Quasiparticle interference (momentum dependent correction to the LDOS)  $\delta\rho(\mathbf{q}, \omega = 0)$  maps shown in the full Brillouin zone for Born limit scattering in the orbital space  $U = 1$  meV. Cuts (b) for  $k_z = 0$  and (c)  $k_z = \pi$ . The sign of the correction to the density of states for the intraband scattering reflects whether the band is hole-like (positive) or electron-like (negative)

for each orbital. The resulting normal state quasiparticle interference (momentum-dependent correction to the local density of states) map is shown at zero frequency in Fig. 2 for the two characteristic  $k_z$  cuts and employing the Born approximation with a weak scattering potential  $U = 1$  meV. Comparing the patterns to the ones found in experiment in Ref. 35, it appears that figure 2(b) is in closest agreement, with well defined intraband ( $q \approx 0$ ) and interband ( $q \approx (\pi, 0)$ ) peaks.<sup>43</sup> The structure of the Fermi surface and the character of the carriers can be easily deduced from the patterns. Once the sign of the impurity potential is fixed (we assume it to be positive for clarity), the scattering within hole or within electron pockets are clearly separable. While the scattering within hole pockets at  $q = 2k_F$  is positive, it is negative for the electron ones. This is determined by the initial sign of electronic dispersion with respect to the chemical potential. The interband scattering between electron and hole pockets contains both positive and negative contributions to  $\delta\rho$ . In particular, there are two processes of opposite signs, see Fig. 2 (a). The first one is hole-like (positive) and another one is electron-like (negative). Observe that if the hole pocket is larger than the electron one, the hole-like scattering can appear in the second BZ, while the electron-like is in the first BZ, and vice versa if the pocket sizes are reversed. Thus the interband peaks in Fig.2(b),(c) can be either positive or negative depending on the relative size and ellipticity of the pockets.

We first examine the QPI peaks that occur due to the complicated Fermi surface in the normal state. The strongest contribution to the LDOS is given by scattering between like orbitals within the large hole pocket (of  $xy$ -character), as well as within the small hole pocket ( $xz, yz$ -character). The interband scattering between electron and hole pockets, again

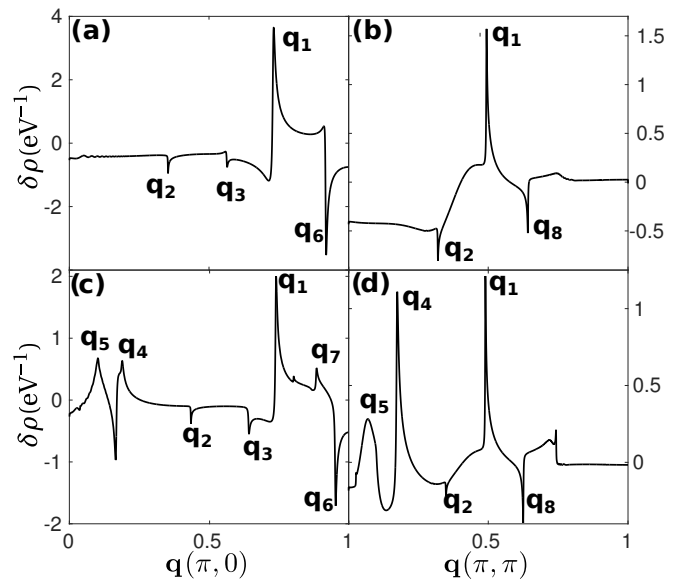


FIG. 3. Cuts taken from figure 2.  $\delta\rho(\mathbf{q}, \omega = 0)$  calculated along the symmetry routes  $\Gamma \rightarrow X$  (left panel) and  $\Gamma \rightarrow M$  (right panel) in the first Brillouin zone for  $k_z = 0$  (a),(b) and  $k_z = \pi$  (c),(d). The corresponding scattering wavevectors at the Fermi surface cuts are shown in figure 1.

of  $xz, yz$ -character (Fig. 1), is also significant. To visualize the corresponding structure of the scattering peaks we present in Fig. 3 the  $\delta\rho(\mathbf{q}, \omega)$  along the  $\Gamma \rightarrow X$  and  $\Gamma \rightarrow M$  high symmetry paths of the first BZ for two different  $k_z$  cuts. Observe that all the scattering events can be clearly identified with processes corresponding to either intraorbital intraband and interband scattering. In particular, as shown in Fig. 1, scattering vectors  $\mathbf{q}_1$  till  $\mathbf{q}_4$  refer to the intra pocket scattering and the amplitude of intra hole and intra electron pocket scattering differs in sign. While the correction to the LDOS from the scattering on the electronic band has a negative sign, the intrapocket scattering for the hole-like bands is positive (for assumed  $V_{imp} > 0$ ; all signs reverse for the opposite case in the weak scattering limit). This difference is related to the overall sign of the electron and hole dispersion. This information can be used to identify interpocket scattering vectors between the pockets of the same character. For example,  $\mathbf{q}_5$  refers to the scattering between the small hole pockets, while  $\mathbf{q}_8$  is the scattering between two electron pockets located at  $X$  and  $Y$  point of the BZ. For interband scattering we identify the scatterings between an electron pocket and the large hole pocket which are labeled as  $\mathbf{q}_6$  and  $\mathbf{q}_7$  by examining the corresponding energy cuts in the first BZ.

Once the scattering events in the Born limit are understood, we should mention that by increasing the impurity scattering strength we find within T-matrix that there are also interorbital scattering components introduced for example between larger and smaller hole pockets, which are however still weaker than the intraorbital ones. In the following, however, we proceed directly to the superconducting state.

### III. SUPERCONDUCTING STATE

Superconducting gap magnitudes and their angular variations on the electron and hole pockets are known for LiFeAs from ARPES<sup>7,30,31</sup> and STM<sup>34</sup> measurements yet their relative phases are unknown. In particular, the magnitude of the superconducting gap for the small hole pockets is  $\Delta_h = 5$  meV, for the large hole pocket  $\Delta_H = 2.5$  meV and for the electron pockets  $\Delta_e \approx 3$  meV. These measurements do not fix the gap structure completely, however, because the relative phases are unknown. There have been a number of theoretical proposals regarding the phase structure of the superconducting gap in LiFeAs. In the original analysis using spin-fluctuation mediated Cooper-pairing performed by Wang et al. 7 using the tight-binding band structure discussed above, the usual  $s_{+-}$ -wave symmetry of the superconducting gap with an overall change of sign between electron and hole pockets was found. On the other hand, the calculated gaps on the small Z-centered pockets were too small compared to experiment. Several subsequent works explored ways to cure this discrepancy<sup>9,13,14</sup>. In Ref.<sup>9</sup>, it was pointed out that once the spin fluctuations are relatively weak, which seems to be the case in LiFeAs<sup>44</sup>, there are several  $s$ -wave channels which are nearly degenerate<sup>9</sup>. Most importantly, these  $s$ -wave states may involve sign changing gap on the two hole pockets<sup>9</sup> as well as orbital-antiphase  $s$ -wave gap<sup>13</sup>, which is stabilized when the pairing interaction is closer to being diagonal in orbital space than band space<sup>8</sup>.

Note that all of these works claim to be roughly consistent with existing ARPES experiments, but find different sign structures of the gaps on the various Fermi surface sheets. In the following, we compute the QPI signatures of each of the  $s$ -wave states, which should help to identify the particular symmetry of the order parameter in LiFeAs. In real STM measurements, electrons from a range of  $k_z$  states are involved in the tunneling process, so no meaningful choice of a single  $k_z$ -cut is possible. In 2D systems like cuprates, the influence of the third momentum dimension is negligible, but in LiFeAs this is not the case (Fig. 1). While electron and large hole pockets correspond to bands that are barely dispersing in the  $k_z$ -direction, the same is not true for the small hole pockets. Nevertheless, empirically the QPI data on LiFeAs seem to contain large scattering vectors that connect the small hole pockets with the electron pockets, suggesting that the  $k_z = \pi$  states play an essential role. With this in mind, we continue our calculation with  $k_z = \pi$  where all pockets are visible on the Fermi surface.<sup>45</sup>

In the superconducting state, Eq. 4 is generalized to

$$\rho(\omega) = -\frac{1}{2\pi} \text{Tr} \text{Im} \sum_{\mathbf{k}} (\tau_0 + \tau_3) \hat{G}^0(\mathbf{k}, \omega) \quad , \quad (5)$$

where  $\tau_0$  and  $\tau_1$  are Pauli matrices in Nambu (particle-hole) space. We show the total density of states in Fig. 4 for the gap structure ( $\Delta_h, \Delta_H, \Delta_e$ ) as determined by ARPES. A typical  $U$ -shaped spectrum is found with all three gaps visible. All the gaps are well pronounced in the spectrum. At energies above the coherence peaks, normal state properties are

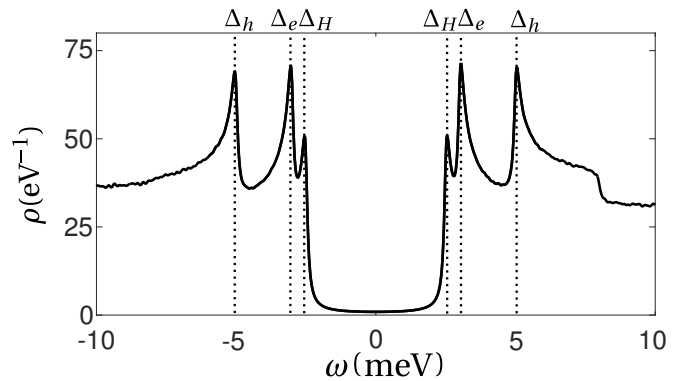


FIG. 4. Density of states in the superconducting phase  $\rho(\omega)$ . Integrated over the full Brillouin zone and in presence of all three hole pockets at  $k_z = \pi$ . Vertical dashed lines indicating gap values  $\Delta_h = 5$  meV,  $\Delta_H = 2.5$  meV and  $\Delta_e = 3$  meV.

recovered, showing a weak particle hole asymmetry. Hence, the antisymmetric part of the correction to the LDOS can be found as

$$\delta\rho^-(\mathbf{q}, \omega) = \text{Tr} \text{Im} \sum_{\mathbf{k}} \tau_3 \hat{G}^0(\mathbf{k}, \omega) \hat{t}(\omega) \hat{G}^0(\mathbf{k} + \mathbf{q}, \omega), \quad (6)$$

where the  $t$ -matrix in the Nambu space is given by

$$\hat{t}(\omega) = [1 - \tau_3 \hat{U} \sum_{\mathbf{k}} \hat{G}(\mathbf{k}, \omega)]^{-1} \tau_3 \hat{U} \quad (7)$$

and  $\tau_3$  is the corresponding component of the Pauli matrix. In the following we will consider various representation of the phases of the superconducting gap on the Fermi surface pockets. However, we will also study the role of the impurity potential in changing the characteristic features of the  $\delta\rho^-(\mathbf{q}, \omega)$ .

We remind the reader of the basic finding of our previous analysis. The peculiar behavior of the antisymmetrized correction to the local density of states due to non-magnetic impurity was evaluated by us previously for the simple model of two parabolic bands (one hole-like and one electron-like) crossing the Fermi level within the T-matrix approximation.<sup>24</sup> It turns out that this quantity contains the term

$$\delta\rho^-(\omega) \propto \text{Im} \frac{\Delta_e \Delta_h}{\sqrt{\omega^2 - \Delta_e^2} \sqrt{\omega^2 - \Delta_h^2}} \quad (8)$$

which is either negative in the frequency interval between  $\Delta_e$  and  $\Delta_h$  if the electron and the hole gaps are out of phase ( $s_{+-}$ ) or is a sign changing function between  $\Delta_e$  and  $\Delta_h$  if the electron and the hole gaps are in phase ( $s_{++}$ ). This correction is the strongest for the large  $\mathbf{q}$  scattering between electron and hole pockets. Therefore, our original proposal in Ref.<sup>24</sup> was based on plotting the interband contribution to the local density of states at large momentum transfer. However, this term also remains clearly observable even if one sums all contributions and for the realistic model system. In particular, we demonstrate below that for the different magnitude superconducting gaps on the hole and the electron pockets, the characteristic behavior of the sign changing gap is also visible in the total  $\delta\rho^-(\omega)$  as is the case for LiFeAs.

### A. $s_{+-}$ -wave gap: impurity potential in orbital and band bases

Before we start our analysis by studying the different possible gap structures, we need to point out the importance of the actual model used for the impurity. First, we consider an impurity which is diagonal in orbital space,  $V_{imp}^{\ell_1 \ell_2} = U \delta_{\ell_1 \ell_2}$ , and consider the effect of the potential strength  $U$  on our ability to identify sign-changing gaps. For simplicity, we first consider the usual  $s^\pm$ -wave superconducting gap *i.e.*, the overall phase of the gap changes from 0 to  $\pi$  between electron and hole pockets. In Fig.5 we show the bias dependence of the full momentum space integrated correction to the LDOS in the Born, intermediate and strong scattering limit in the presence of all three hole pockets obtained by

$$\delta\rho^-(\omega) = \sum_{\mathbf{q} \in \Omega} \delta\rho^-(\mathbf{q}, \omega), \quad (9)$$

where the integration is over the entire momentum space, or, in practical application, over a well defined area  $\Omega$  around the scattering peaks<sup>24</sup>.

The peculiar behavior of the total  $\delta\rho^-(\omega)$  summed over all  $\mathbf{q}$  wave vectors provides valuable information on whether the gap is a sign changing function itself without addressing the actual symmetry or detailed structure of the gap. This is essentially because the contributions arising from wave vectors that connect bands with order parameters of the same sign have negligible amplitude compared to those involving sign changes<sup>24</sup>. This knowledge is important, especially if it turns out to be difficult experimentally to address the actual symmetry of the gap within QPI, since the sign-changing gap function is usually a strong hint in favor of unconventional superconductivity. Thus, even the general statement on the sign-changing gap signatures observed from the antisymmetrized correction to the local density is of general interest.

Comparing the Born limit with  $U = 1$  meV shown in Fig.5(b) to the result of the intermediate strength scattering  $U = 10$  meV shown in Fig. reffig:Fullqint(a), we find them in close agreement with each other. The clear “even” behavior of  $\delta\rho^-(\omega)$  for the sign changing  $s$ -wave state and the “odd” one for the  $s_{++}$  state are expected.<sup>24</sup> Here we use the imprecise terms “even” and “odd” behavior of the antisymmetrized LDOS to refer, respectively, to single-sign, resonant behavior of  $\delta\rho^-$  as a function of bias between the gap energies for the  $s_{+-}$  state and the weaker response with a zero-crossing for the  $s_{++}$  state.<sup>24</sup>

The characteristic signal changes as soon as we introduce stronger scattering amplitudes  $U = 0.2$ eV in Fig. 5(c). An in-gap bound state develops for  $s_{+-}$  symmetry, moving from the gap edge toward smaller energies with increasing scattering strength. The occurrence of in-gap bound states is of course itself a strong hint toward a sign change of the order parameter; nevertheless the distinction between even and odd  $\delta\rho^-$  signals in the bias region between the two gap energies remains unambiguous, so the antisymmetrized LDOS analysis may also be used as a test of gap sign change after subtracting any bound states<sup>24,29</sup>.

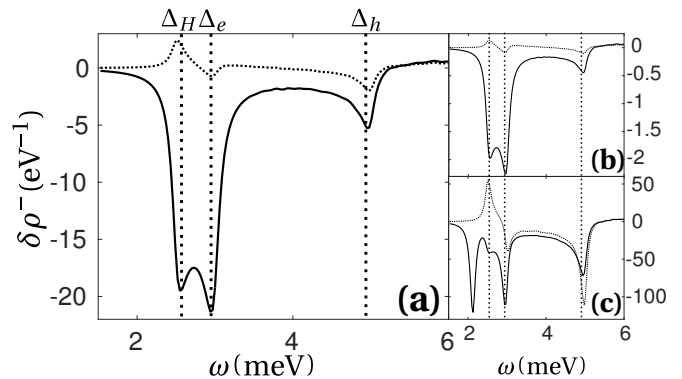


FIG. 5. Antisymmetrized  $\mathbf{q}$  integrated correction to the LDOS  $\delta\rho^-(\omega)$  for non-magnetic impurity scattering diagonal in orbital space. (a) intermediate limit full  $t$ -matrix calculation  $U = 10$  meV, (b) Born approximation  $U = 1$  meV and (c) unitary limit full  $t$ -matrix calculation  $U = 0.2$ eV integrated over the full Brillouin zone and in presence of all three hole pockets at  $k_z = \pi$ . Vertical dashed lines indicating gap values. Result is shown with solid lines for conventional  $s_{+-}$ , dashes lines for  $s_{++}$ .

While for a general statement of the sign change this is sufficient, the distribution of signs over the different pockets is not determined by this procedure. LiFeAs, with its well studied Fermi surface, is a promising material for addressing possible scattering events separately. In order to use the known fermiology to restrict our integration range to a well defined area of  $\mathbf{q}$ -space and study the response in an experimentally relevant case, we consider scattering in band space, where we can select scattering events simply by including or excluding specific pockets in the scattering and allowing all intra and interband scattering with similar magnitudes. This form of impurity, when transformed to orbital space, corresponds to a non-local impurity with strong interorbital scattering. Most importantly, an inclusion of the non-local corrections to the impurity potential results in new scattering wave vectors (e.g., the one between inner and outer hole pockets, absent for the purely local impurity defined in orbital space) and different distribution of the intensity among them.

In particular, in Fig.6(a) the  $q$  integrated scattering is shown in the Born limit. Choosing  $U_{inter} = U_{intra} = 1$  meV, we find a similar behavior for the  $s_{++}$  and  $s_{+-}$  case to those shown previously in Fig.5 for the impurity potential defined in orbital space. This indicates that the behavior is universal and does not depend on the way the impurity potential is introduced.

In realistic STM experiments, the measurements are done at finite yet very low temperatures. As we have shown previously in Ref. 24, a thermal broadening smears out the clear peak structure with increase of temperature. Nevertheless, as we show in Fig. 7, the characteristic features of the  $s_{+-}$  and  $s_{++}$ -states remains still visible up to  $T = 0.6T_c$ . Here, the temperature dependence is introduced using BCS-type behavior of the gap magnitudes on each Fermi surface pocket and thermally averaging the antisymmetrized correction to the lo-

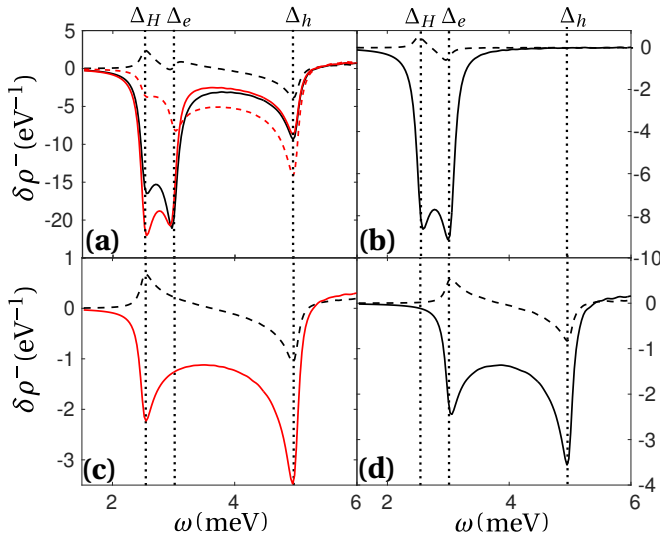


FIG. 6. Antisymmetrized  $\mathbf{q}$  integrated correction to the LDOS  $\delta\rho^-(\omega)$  for non-magnetic impurity scattering potential diagonal in the band space and in the Born limit  $U_{\text{inter}} = U_{\text{intra}} = 1$  meV. (a) sum of all inter and intraband scatterings, (b) integrated  $q$  area containing large hole and electron pocket scattering, (c) integrated  $q$  area containing large hole and small hole pocket scattering and (d) small hole and electron pockets scattering. Dashed vertical lines indicate the magnitudes of the corresponding gaps. Solid black curves refer to the conventional  $s_{+-}$ -state, with all hole pocket gap signs the same, and opposite to that on the electron pockets (Fig. 8(B)). The dashed black curves correspond to  $s_{++}$ , the solid red line refer to the state containing a sign change of the gap on the large hole pocket Fig. 8(C), while the dashed red one corresponds to the sign change on small hole pockets (Fig. 8(D)).

cal density of states

$$\langle\delta\rho^-(\Omega)\rangle \equiv \int_{-\infty}^{\infty} d\omega \delta\rho^-(\omega) \left[ \frac{-\partial f}{\partial\omega}(\omega + \Omega) - \frac{-\partial f}{\partial\omega}(\omega - \Omega) \right]$$

### B. Other possible $A_{1g}$ -symmetry states with sign-change gap between hole pockets

Due to the relative weakness of the spin fluctuations at the wave vector corresponding to the scattering between electron and hole pockets in LiFeAs<sup>46</sup>, it has been proposed<sup>9</sup> that the distribution of the phases of the superconducting gap magnitudes on the Fermi surface sheets could be different than it is in the conventional  $s_{+-}$ -wave scenario. Overall the gap would still possess the global  $A_{1g}$ -symmetry, yet the phase structure could be richer. Some such states are depicted in Fig.8.

The natural question arises whether QPI could be used to determine these additional phase shifts. We first note that the  $A$ -state has an additional sign change between two tiny hole pockets, located near the  $\Gamma$ -point. For the QPI it would mean to search for the characteristic features in  $\delta\rho^-(\omega)$  for small scattering wavevectors ( $\mathbf{q}_4$  and  $\mathbf{q}_5$ ), which is in general difficult for the QPI experiments as it requires a Fourier transform

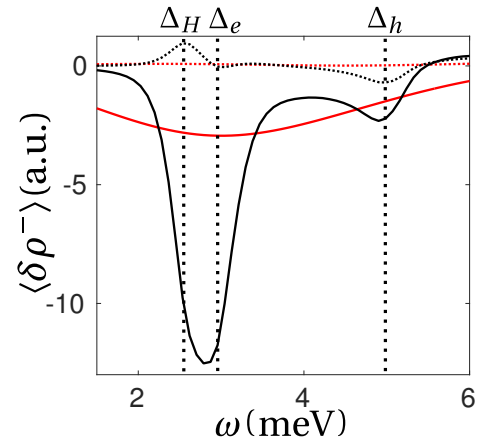


FIG. 7. Thermally averaged antisymmetrized full- $\mathbf{q}$  integrated LDOS change  $\langle\delta\rho^-(\omega)\rangle$  for nonmagnetic scattering in the intermediate limit  $U = 10$  meV (the  $\delta\rho^-(\omega)$  is shown in Fig.5(a) and  $k_z = \pi$ ). The black and red curves refer to  $T = 0.2T_c$  and  $T = 0.6T_c$ , respectively. Solid curves refer to the  $s_{+-}$  state, while the dashed ones for conventional  $s_{++}$  state.

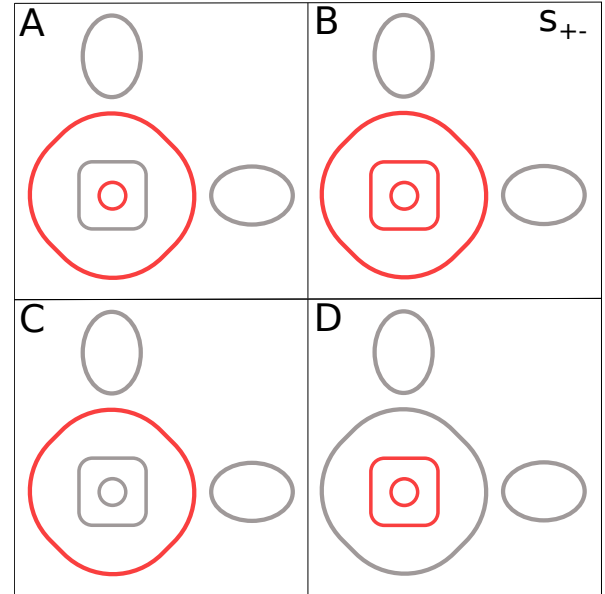


FIG. 8. Possible sign-changing states within  $A_{1g}$ -symmetry representation found previously in Ref.<sup>9</sup>. The conventional  $s_{+-}$ -state is referred as  $B$ -state. Red and grey indicate different signs of the gap function on the Fermi surface. Note we do not discuss the interesting further possibility that there might be complex phases among the gap functions on the different Fermi sheets, as predicted in some cases<sup>15</sup>. This question has been addressed in Ref. 47, where it was found that such complex states did not yield clear qualitative signatures in  $\delta\rho^-$ .

of Friedel oscillations from an isolated impurity recorded in a large field of view in the real space. In addition these small pockets exist only near  $k_z = \pi$  and have much smaller densities of states, which represents an additional problem. Therefore, we first address the question of whether some less subtle possible sign changes beyond the usual  $s_{+-}$  picture can be still detected. We plot in Fig.6(a) the results for the two other

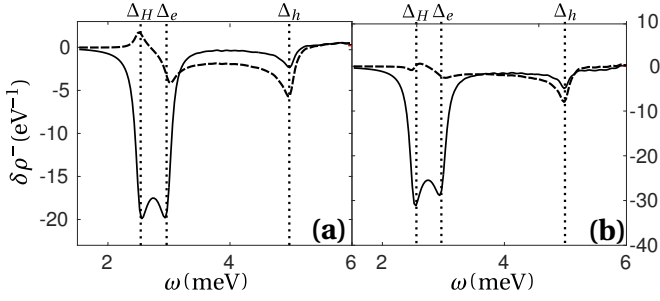


FIG. 9. Antisymmetrized  $\mathbf{q}$  integrated correction to the LDOS  $\delta\rho^-(\omega)$  for the  $C$  and  $D$ -states, depicted in Fig.8, using the non-magnetic impurity diagonal in orbital space using  $t$ -matrix calculation for  $U = 10$  meV in presence of all three hole pockets at  $k_z = \pi$ . (a) refers to full  $\mathbf{q}$  integration of the QPI map as also shown Fig.5(a). (b) shows  $q$ -selected scattering around  $q_7$ . Solid curves refer to the  $C$ -state (sign change on the large hole pocket), while the dashed curves refer to the  $D$ -state.

candidate states for LiFeAs, namely for the superconducting state where the large hole pocket of mostly  $xy$ -character has opposite sign to all other electron and hole pockets ( $C$ -state) and the one where the tiny inner hole pockets have opposite sign to the large hole and electron pockets ( $D$ -state). We find an even response for the total integrated  $\delta\rho^-(\omega)$ , for each sign-changing gap structure, when the impurity scattering potential is written in the band basis. Thus, it appears that the magnitude of the response at the different gap values provides direct information regarding the pockets in which the sign change occurs. To specify this further we show in Fig.6 (c)-(d) the integrated responses around each corresponding  $q$  vector i. e. large hole to electron, small hole to electron and hole to hole scattering, respectively. All three possible scattering events show indeed the expected even or odd symmetry when the change of the sign of the gap occurs. Here, we point out that the inter-pocket scattering between inner and outer hole pockets is not always present, which is related to the structure of the impurity potential in the orbital and band basis and the fact that inner and outer pocket have different orbital character ( $xz/yz$  for the inner one and mostly  $xy$  for the outer one). These scattering wave vectors, if observed, could play an important role in determining the overall structure of the superconducting gap in LiFeAs.

In this regard, to see whether  $C$  and  $D$  states can be identified, we now return to a more realistic situation, when the impurity potential is nearly diagonal in the orbital basis. This seems to agree better with ab-initio calculations<sup>39,42</sup>. The problem is further complicated by the fact that beyond the weak scattering (Born) limit, the  $t$ -matrix introduces additional interorbital scattering terms. Here we study the effects of such terms by considering an intermediate strength scatterer  $U = 10$  meV that does not create bound states in the gap. In Fig.9 (a) we show the antisymmetrized response integrated over the full  $q$  space for the  $C$  and the  $D$ -states. We remind the reader that for the  $s_{++}$  and the conventional  $s_{+-}$  states, the results are clearly separable independent of the way the impurity potential is written. Regarding the more complicated  $C$ - and the  $D$ -states, we note that the relative sign change ( $C$ -state) or its absence ( $D$ -state) between large hole and electron

pockets, i.e. between  $\Delta_H = 2.5$  meV and  $\Delta_e = 3$  meV is clearly visible. At the same time, the variation of  $\delta\rho^-$  between smaller gaps,  $\Delta_e$  and the large gap,  $\Delta_h = 5$  meV, on the small pockets is trickier due to a more complex phase structure of the overall gap. While for the  $D$ -state the clear sign-changing behavior is indeed observed, as expected, the behavior of  $\delta\rho^-$  cannot be clearly assigned for the  $C$ -state. In the latter state,  $\Delta_e$  has opposite sign to  $\Delta_H$ , but the same as on  $\Delta_h$ , so the behavior of  $\delta\rho^-(\omega)$  loses its characteristic behavior between  $\Delta_e$  and  $\Delta_H$ . The situation does not really improve if one selects some specific  $q$ -scattering wavevector such as  $q_7$ , which is responsible for the scattering between small hole pockets and electron pockets. The results in Fig. 9 (b) again show a clear signature of the phase structure between  $\Delta_H$  and  $\Delta_e$ , but cannot be unambiguously assigned for frequency region between  $\Delta_e$  and  $\Delta_h$ .

The result of this exercise shows that the determination of the superconducting gap structure on the multiple Fermi surface sheets may be a challenging task if gaps change sign more than once (i.e., a frustrated case), or if some gaps are close in magnitude. For this situation,  $\delta\rho^-(\omega)$  can be still helpful to identify whether the gap is generally sign changing at least on some of the pockets, however, the overall phase structure distribution over the multiple sheets cannot be unambiguously determined.

### C. Orbital antiphase superconductivity

Another very interesting proposal put forward for LiFeAs is the so-called orbital-antiphase superconducting gap<sup>8</sup>, originally proposed for the iron-based superconductors within an effective model<sup>48</sup>. In this essentially strong-coupling approach, the phase structure of the superconducting gap is defined not on the Fermi surfaces but on the orbitals. This scenario appears plausible for LiFeAs due to the strong orbital differentiation of the Fermi surface sheets. In particular, we present the Fermi surface cuts for  $k_z = \pi$  with orbitally-resolved matrix elements in Fig. 10. For example, the  $C$ -state discussed in the previous subsection can also be regarded as a type of orbital antiphase state because the superconducting gap on the large hole pocket, which has mostly  $xy$ -orbital character, has opposite sign to the other pockets, which mostly consist of the hybridized  $xz, yz$ - and also  $xy$ -orbitals. Nevertheless, the  $C$ -state still appears within the solution of the BCS-type of equations in the band basis, assuming the superconductivity arises as an instability of the electron gas with respect to the actual Fermi surface. Therefore, assuming a sign-changing but otherwise constant gap on the orbitals must result in a rather strong angular dependence of the superconducting gap on the Fermi surface. The question to ask then is whether such a sign change between orbitals can be identified in QPI experiments. To do so, we adopted the scheme in which we introduce the superconducting gaps in the orbital representation before the unitary transformation from the orbital to the band space is performed. For the sake of simplicity we took the orbital analog of the  $C$ -state and assumed that the sign of the gap on the  $xy$ -orbital is either opposite or the same



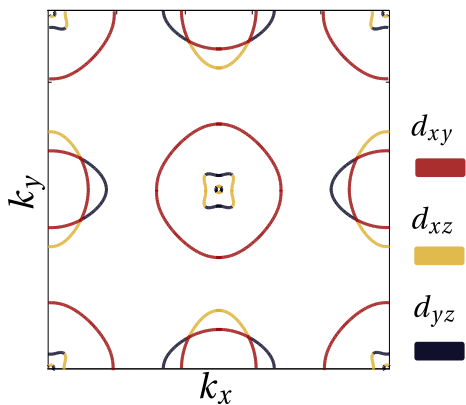


FIG. 10. Orbitally-resolved Fermi surface cuts for  $k_z = \pi$ . The contribution of the three dominant orbitals  $d_{xy}$ ,  $d_{xz}$ ,  $d_{yz}$  is shown.

to the signs of the gaps on the  $xz, yz$  orbitals. As the other two orbitals  $x^2 - y^2$  and  $3z^2 - r^2$  do not contribute strongly to the states at the Fermi level, we assumed their gaps to be small and their phase structure irrelevant. In particular, we choose  $\Delta_{xy} = 3.2$  meV and  $\Delta_{xz} = \Delta_{yz} = 5$  meV, which produce the sizes of the superconducting gaps on the actual Fermi surface sheets roughly consistent with ARPES experiments.

In particular, in Fig. 11 we show the resulting gap structure on the Fermi surface for the  $k_z = \pi$  cuts for the orbital in-phase (a) and antiphase (b) superconducting gap. We find clearly anisotropic gaps on each Fermi surface sheet in both cases. Nevertheless, the anisotropy is much stronger for the  $s_{+-}$ -wave case. In particular, the strongest anisotropy occurs on the electron pockets, with maximum gap values on the tips of the elliptical pockets and deep minima or accidental nodes near the intersection regions. Also consistent with the ARPES experiments,<sup>30</sup> the largest gap occurs on the tiny hole pockets. The smallest gaps occur on the inner part of the elliptic electron pockets and on the large hole pocket. Overall, the gap structure and its anisotropy on the Fermi surface pockets is roughly consistent with experimental data<sup>7,30,31,34</sup>, except for the existence of nodes or deep minima near the intersections of the two electron pockets.

In a microscopic spin fluctuation theory assuming an LDA+DMFT electronic structure and self-energy, a pairing interaction very close to the phenomenological orbital-diagonal pairing vertex assumed here was calculated.<sup>8</sup> However in that work the Fermi surface did not agree with ARPES, and although the electron pockets were found to be fully gapped, they were still considerably more anisotropic than found in experiment. Furthermore, the electronic structure used in that work was not consistent with the glide plane symmetry of the LiFeAs crystal; when some of the same authors redid the calculation incorporating this symmetry<sup>49</sup>, they found sizeable interorbital pairing matrix elements, and a conventional  $B$ -type ground state as in Ref. 7.

In Fig. 12, we also show the calculated density of states for the orbital antiphase and in-phase order parameter, respectively. Observe that the angular dependence of the gap magnitudes on the Fermi surface is reflected in the low-energy

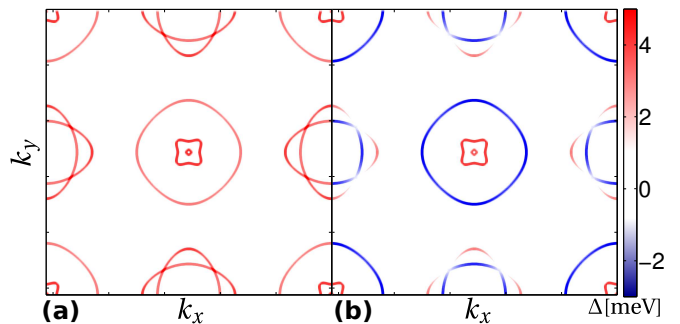


FIG. 11. The resulting gap structure on the Fermi surface for the  $k_z = \pi$  cuts for the orbital in-phase (a) and antiphase (b) superconducting gap, depicted for the LiFeAs Fermi surface for  $k_z = \pi$ . The superconducting gaps  $\Delta$  in orbital space are projected by unitary transformation onto the band gaps on the corresponding Fermi surface sheets. Here, the effect of the interband gaps is neglected as it appears to be negligibly small on the Fermi surface even near the crossing points of the two electron pockets. The resulting gap resembles the so-called  $C$ -phase, except for the nodes and the sign changes in the electron pockets.

behavior of the density of states. While the  $s_{++}$  orbital in-phase gap shows nodeless behavior at low energies, the orbital antiphase  $s_{+-}$  results in a near-nodal power-law behavior of the density of states. Next we study this more complex

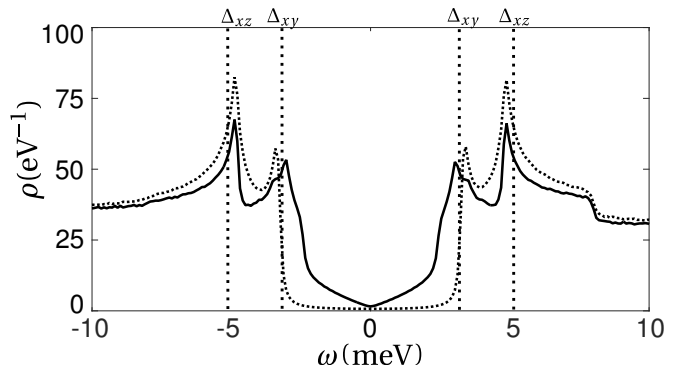


FIG. 12. Calculated density of states for the orbital-antiphase ( $s_{+-}$ -solid curve) and orbital in-phase ( $s_{++}$ -dashed curve) superconducting gap,  $\rho(\omega)$ , using gap values  $\Delta_{xy} = 3.2$  meV,  $\Delta_{xz} = \Delta_{yz} = 5$  meV.

gap structure in terms of the correction to the LDOS due to a non-magnetic impurity diagonal in orbital space. As we mentioned earlier, the determination of the phase structure of the superconducting gap on the Fermi surface appears to be a non-trivial task due to both non-zero intra- and interband gaps and strong orbital mixing on some Fermi surface sheets. Therefore, to determine whether or not the gap changes sign on various orbitals and not necessarily between the bands is a challenging task. We show in Fig. 13 the momentum integrated antisymmetrized correction to the density of states,  $\delta\rho^-(\omega)$ , in the intermediate impurity scattering limit,  $U = 10$  meV, for the orbital in-phase  $s_{++}$ -state, and orbital antiphase  $s_{+-}$ -state, respectively. Not surprisingly, we still find a clear signature of the sign-changing gap despite the fact that the gaps

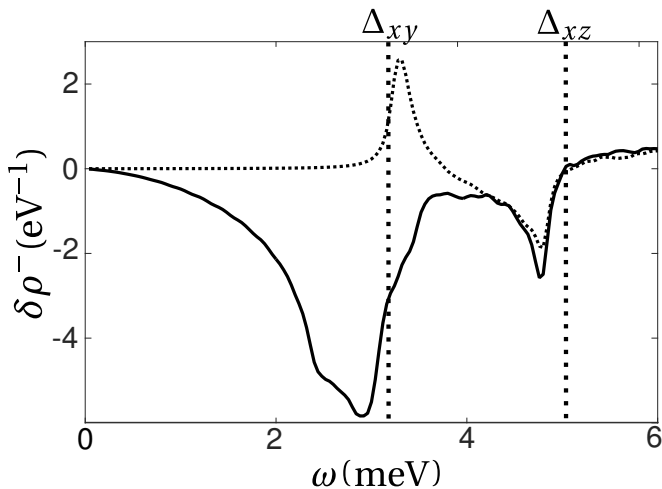


FIG. 13. Momentum-integrated bias-dependence of the antisymmetrized correction to the LDOS,  $\delta\rho^-(\omega)$  for LiFeAs at  $k_z = \pi$ . Here, we use  $\Delta_{xy} = \pm 3.2$  meV and  $\Delta_{xz} = \Delta_{yz} = 5$  meV. The solid curve refers to the orbital-antiphase  $s_{+-}$ , while the dashed curve is for orbital-in-phase gap,  $s_{++}$ .

reside on the orbitals rather than on bands, but their phase structure, especially on the electron pockets, cannot be clearly defined. Nevertheless, the behavior of the  $\delta\rho^-(\omega)$  within the energy region of interest (*i.e.* between 3 and 5 meV) is still very characteristic. At the same time, the position of the peaks is less informative in this case as it refers to the maximum of the gaps on the bands. As a result, using QPI in this case again only allows one to determine the presence of the sign change in the gap structure, but not the precise distribution of it among the various bands.

#### IV. CONCLUSION

We have investigated an extension of the phase-sensitive method of analyzing QPI data from non-magnetic impurities, proposed by us previously<sup>24</sup> in the context of a simplified model, for a realistic 10-orbital tight-binding Hamiltonian and  $t$ -matrix approximation for the impurity potential. We have concentrated on studying the LiFeAs compound due to a rather detailed experimental knowledge of the electronic structure of this system<sup>7</sup> and the measured superconducting gap values for each of the Fermi surface sheets<sup>30,31</sup>. In particular, we have shown that despite the complex Fermi surface topology, the Friedel oscillations originating from various intraband and interband impurity-induced scattering can be clearly separated in LiFeAs. Furthermore, the scattering within hole or electron pockets as well as between the electron and hole Fermi surface pockets show some characteristic features, which should help to identify them experimentally. Furthermore, the behavior of the phase-sensitive antisymmetrized correction to the local density of states,  $\delta\rho(\omega)$ , in the superconducting state allows one to discriminate between the sign-changing and sign-preserving superconducting gaps indepen-

dent of the structure and the strength of the impurity potential. In particular, we showed that the relative phase difference of the gap between large hole pocket and electron pockets can be easily addressed within QPI experiments.

At the same time, the determination of the relative phase of the gap on the tiny  $Z$ -centered hole pockets with respect to the other electron and hole bands appears to be a much more subtle issue. It is still possible to distinguish between a conventional constant sign gap and the usual  $s_{+-}$  scenario where the gap is one sign on all hole pockets and another on the electron pockets. However, once the sign structure of the gap is more complex and involve further sign changes between large hole and small hole pockets, the behavior of  $\delta\rho^-(\mathbf{q}, \omega)$  cannot be clearly assigned, primarily due to the near proximity of two gap energies in this system. We discussed the same point for the so-called orbital-antiphase superconducting gap structure, where the gap is diagonal in orbital space, raised in the context of the LiFeAs system<sup>8</sup>.

We emphasize again that our primary goal was to obtain qualitative statements about sign change in the superconducting gap structure. It is interesting to note that the momentum dependence of the effective impurity potential appears to be irrelevant for the simplest weak scattering calculation of the antisymmetric fully  $\mathbf{q}$ -integrated QPI signal, as can be seen by an expansion of the  $T$ -matrix in harmonics. Although this is only approximately true for a realistic system with anisotropic gaps, etc., and it does not hold for restricted  $\mathbf{q}$ -integrated QPI intensities, we expect that our basic conclusions are only weakly affected by any momentum dependence induced by the unitary transformation of the bare potential or, for example, by antiferromagnetic correlations.

In summary, the method we proposed seems to allow one to answer the question whether the gap has a sign change, but cannot be used, at least in LiFeAs, for a more detailed determination of the phase structure of the order parameter.

*Note added.* In the final stages of writing, two papers appeared that discussed detailed analysis of quasiparticle interference measurements on LiFeAs, including a new qualitative phase-sensitive method based on interference of impurity bound states<sup>26,27</sup>. In Ref. 27, the authors explicitly perform an analysis of the antisymmetrized  $\mathbf{q}$ -integrated differential conductance according to the prescription presented in Ref. 24 and here. While they point out a substantial quantitative dependence on the area in  $\mathbf{q}$  space over which one chooses to integrate, the qualitative result is of the “even” type between the two gap energies, in the nomenclature of the current paper. Thus their results using our method are entirely consistent with the results they obtain using their bound state approach, and with the claim already made in Ref. 24, namely that there is a sign change between hole and electron pockets in LiFeAs.

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