

**Effect of nonsymmorphic space groups on correlation functions in iron-based superconductors**R. Nourafkan<sup>1</sup> and A.-M. S. Tremblay<sup>1,2</sup><sup>1</sup>*Département de Physique, Institut quantique and Regroupement québécois sur les matériaux de pointe, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K 2R1*<sup>2</sup>*Quantum Materials Program, Canadian Institute for Advanced Research, Toronto, Ontario, Canada M5G 1Z8*  
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The orbital basis is natural when one needs to calculate the effect of local interactions or to unravel the role of orbital physics in the response to external probes. In systems with nonsymmorphic point groups, such as the iron-based superconductors, we show that symmetries that emerge in observable response functions at certain wave vectors are absent from generalized susceptibilities calculated with tight-binding Hamiltonians in the orbital basis. Such symmetries are recovered only when the generalized susceptibilities are embedded back to the continuum using appropriate matrix elements between basis states. This is illustrated with the case of LiFeAs and is further clarified using a minimal tight-binding Hamiltonian with nonsymmorphic space group.

DOI: [10.1103/PhysRevB.96.125140](https://doi.org/10.1103/PhysRevB.96.125140)**I. INTRODUCTION**

A density or magnetic response function measures the response to an applied conjugate field. The density response function, for example, is given by  $\langle \hat{n}(\mathbf{r}, t) \hat{n}(\mathbf{r}', 0) \rangle$ , where  $\hat{n}$  denotes the density operator. Interactions between electrons cause correlations that complicate the calculation of response functions: the electron and the hole created by an external field interact not only separately with the other particles but also with each other. Since correlations affect different orbitals differently, developing a microscopic understanding requires expressing the response function in a suitable orbital basis and studying its different components. How these different components are transformed under point-group symmetry operations is one of the questions that arises naturally.

The symmetry properties of the observable response function is usually discussed in the long wavelength (small wave-vector) limit where the inner structure of the unit cell becomes irrelevant [1]. However, a large number of recent experiments are done at finite wave vectors that probe the inner structure of the unit cell. Furthermore, all approximations for interaction effects are formulated in an atomiclike orbital basis where interaction strengths can be defined more straightforwardly than in the extended Bloch states. Therefore, it is important to verify how the underlying lattice influences the symmetry properties of the correlation functions calculated in the orbital basis.

One of our main messages is that for nonsymmorphic crystals it is necessary to map back the model to the continuum to recover all the symmetries of observable susceptibilities. The generalized susceptibilities in orbital space have a lower symmetry. That result is very general. But we find it preferable to include a specific example. We chose the iron-based superconductors (FeSCs) which are still actively studied. (For a review see Ref. [2].) Similar to the copper oxide high temperature superconductors, these are layered materials where the superconductivity arises in proximity to long-range magnetic order. However, the nature of the magnetic orders differs between the two families. In FeSCs the magnetic order is a stripe-type antiferromagnetic (AF) which lowers the  $S_4$  symmetry (the fourfold rotational symmetry,  $C_4$ , around an Fe

site followed by reflection about the plane) to  $C_2$ . This phase is necessarily accompanied by a structural transition (tetragonal to orthorhombic).

The FeSCs crystalize in a nonsymmorphic space group. Numerous studies have investigated the impacts of the nonsymmorphic space group of FeSCs on their electronic properties in the normal or superconducting phases [3–11]. They either discuss the symmetry of the Bloch states at high symmetry points of the reciprocal lattice or consider possible unconventional Cooper pairing. We instead consider explicit symmetries of the generalized susceptibilities and of the observable susceptibilities in the orbital basis which, to our knowledge, have not been discussed so far. Our goal in this paper is to present a detailed discussion of these symmetries.

One of the nonsymmorphic symmetry operations of the FeSCs is a  $\pi/2$  rotation about an axis perpendicular to the iron-pnictide planes (and centered in the middle of the bond separating two irons) followed by a half-integer translation (see Fig. 1). We show in this paper that the above nonsymmorphic symmetry operation leads, as expected, to generalized correlation functions that are *not* invariant under  $\pi/2$  rotation alone when calculated with a model Hamiltonian in the orbital basis since rotation by  $\pi/2$  alone is not a symmetry. There is however an apparent paradox since correlation functions evaluated at wave vector  $q_z = 0$  should be invariant under  $\pi/2$  rotation, as follows from the fact that rotation by  $\pi/2$  around an iron atom in the  $xy$  plane, followed by a reflection  $z \rightarrow -z$ , leaves the lattice invariant. Here we solve this paradox by showing that to recover the  $\pi/2$  symmetry at  $q_z = 0$ , one must take into account overlap matrix elements between local basis states. These are necessary to embed the orbital-basis model back to the continuum. We call these matrix elements oscillator matrix elements. Note that similar considerations apply to lattices whose space group is symmorphic when a nonprimitive cell is chosen. For these lattices, however, a primitive cell can always be chosen so that the unavoidable complications that we face in the nonsymmorphic case can be avoided in the symmorphic case.

The rest of this paper is organized as follows. In Sec. II we recall the procedure to obtain the Hamiltonian in the orbital

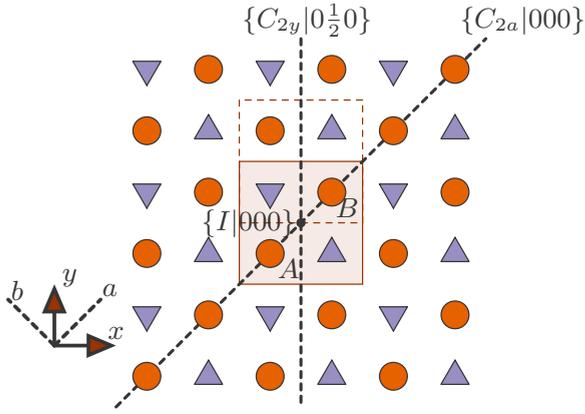


FIG. 1. Sketch of the lattice structure of a trilayer Fe-As unit. The iron and the pnictide atoms are shown respectively by circles and triangles. Down triangles indicate that the As is below the plane and up triangles that As is above the plane. The inversion center is located in the middle of an Fe-Fe link. The three generators of the  $P4/nmm$  space group,  $\{C_{2y}|0\frac{1}{2}0\}$ ,  $\{C_{2a}|000\}$ , and inversion  $\{I|000\}$ , are illustrated. The dashed square shows how the unit cell in the shaded area transforms under  $\{C_{2y}|0\frac{1}{2}0\}$ .

basis. Section III introduces the generalized charge susceptibilities that are usually calculated in the orbital basis and their link to observable charge susceptibilities in the continuum through embedding with oscillator matrix elements. Then we explain in Sec. IV how the orbital-basis Hamiltonian transforms under nonsymmorphic operations. The discussions in these sections are general. Section V presents some results on the bare susceptibility of LiFeAs. Section VI contains concluding remarks. Appendix A demonstrates how oscillator matrix elements appear in the two-body interaction in the orbital basis, Appendix B gives group-symmetry details of LiFeAs, and Appendix C explicitly shows how extra phases appear in the orbital basis because of nonsymmorphic transformations. These transformations are further explained in Appendix D using a minimal tight-binding Hamiltonian to explicitly show that even though  $q_z = 0$  eigenenergies are invariant under  $\pi/2$  rotation, eigenvectors are not. We also show explicitly in this Appendix how oscillator matrix elements allow us to recover  $q_z = 0$  symmetry of the charge fluctuations.

## II. HAMILTONIAN IN THE ORBITAL BASIS

In this section we recall how to write the Hamiltonian in the orbital basis. This allows us to introduce the notation. We emphasize that the discussions in this section and the following section are general. The noninteracting part of the Hamiltonian is given by

$$\hat{\mathcal{H}} = \sum_{\sigma} \int d\mathbf{r} \hat{\Psi}_{\sigma}^{\dagger}(\mathbf{r}) \left[ -\frac{\hbar^2}{2m} \nabla^2 + \sum_{\mathbf{R}_i} V(\mathbf{r} - \mathbf{R}_i) \right] \hat{\Psi}_{\sigma}(\mathbf{r}), \quad (1)$$

where  $\hat{\Psi}_{\sigma}(\mathbf{r}, \tau)$  is the field operator,  $V(\mathbf{r} - \mathbf{R}_i)$  denotes the ionic potential, and  $\mathbf{R}_i$  denotes the lattice sites. The field operators can be expanded in a basis set. In a weakly correlated crystalline system, the prime candidate for the basis set is

Bloch functions. However, in an interacting system it is more suitable to expand the field operators in terms of an atomiclike orbital basis set  $\{\phi_{l\mathbf{R}_i}(\mathbf{r})\}$ , namely,

$$\hat{\Psi}_{\sigma}(\mathbf{r}) = \sum_{l\mathbf{R}_i} \phi_{l\mathbf{R}_i, \sigma}(\mathbf{r}) \hat{c}_{l\mathbf{R}_i, \sigma}, \quad (2)$$

where  $\hat{c}$  denotes the second quantized destruction operator for the state described by the atomic position  $\mathbf{R}_i$  and by the index  $l \equiv (\alpha, l^{\alpha})$  that combines ion (sublattice),  $\alpha$ , and orbital indices  $l^{\alpha}$ . We assume that the basis set is an orthonormal Wannier basis with zero direct overlap between different lattice sites,

$$\int d\mathbf{r} \phi_{l_1\mathbf{R}_1, \sigma}^*(\mathbf{r}) \phi_{l_2\mathbf{R}_2, \sigma'}(\mathbf{r}) = \delta_{\sigma, \sigma'} \delta_{l_1 l_2} \delta_{\mathbf{R}_1, \mathbf{R}_2}. \quad (3)$$

Therefore, the single-particle Hamiltonian can be written as

$$\begin{aligned} \hat{\mathcal{H}} &= \sum_{\sigma} \sum_{l_1 l_2} \sum_{\mathbf{R}_1, \mathbf{R}_2} \mathbf{H}_{\mathbf{R}_1, \mathbf{R}_2}^{l_1 l_2, \sigma} \hat{c}_{l_1\mathbf{R}_1, \sigma}^{\dagger} \hat{c}_{l_2\mathbf{R}_2, \sigma}, \\ \mathbf{H}_{\mathbf{R}_1, \mathbf{R}_2}^{l_1 l_2, \sigma} &= \int d\mathbf{r} \phi_{l_1\mathbf{R}_1, \sigma}^*(\mathbf{r}) \\ &\quad \times \left[ -\frac{\hbar^2}{2m} \nabla^2 + \sum_{\mathbf{R}_i} V(\mathbf{r} - \mathbf{R}_i) \right] \phi_{l_2\mathbf{R}_2, \sigma}(\mathbf{r}). \end{aligned} \quad (4)$$

The two commonly used gauges for the periodic orbital basis are the standard gauge where

$$\phi_{l\mathbf{k}, \sigma} = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_i} \exp(i\mathbf{k} \cdot \mathbf{R}_i) \phi_{l\mathbf{R}_i, \sigma}, \quad (5)$$

with  $N$  the number of unit cells, and the alternative gauge where

$$\phi_{\alpha l^{\alpha} \mathbf{k}, \sigma} = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_i} \exp[i\mathbf{k} \cdot (\mathbf{R}_i + \mathbf{r}_{\alpha})] \phi_{\alpha l^{\alpha} \mathbf{R}_i, \sigma}, \quad (6)$$

with  $\mathbf{r}_{\alpha}$  the position of atom  $\alpha$  within a unit cell.

In the standard gauge, the basis functions are strictly periodic in the Brillouin zone (BZ); hence  $\mathbf{H}_{\mathbf{k}}$  is BZ periodic, i.e.,  $\mathbf{H}_{\mathbf{k}+\mathbf{G}} = \mathbf{H}_{\mathbf{k}}$ , where  $\mathbf{G}$  is a primitive reciprocal lattice vector. In the alternative gauge, the Hamiltonian is given by  $\mathcal{H} = \sum_{\mathbf{k}} \phi_{\mathbf{k}}^{\dagger} \mathbf{H}'_{\mathbf{k}} \phi_{\mathbf{k}}$ , where  $\phi'_{\mathbf{k}} = \mathbf{V}_{\mathbf{k}} \phi_{\mathbf{k}}$ , and  $\mathbf{H}'_{\mathbf{k}} = \mathbf{V}_{\mathbf{k}}^{\dagger} \mathbf{H}_{\mathbf{k}} \mathbf{V}_{\mathbf{k}}$ , where  $\mathbf{V}_{\mathbf{k}}$  is a diagonal unitary matrix with diagonal elements  $\exp(-i\mathbf{k} \cdot \mathbf{r}_{\alpha})$  and  $\mathbf{H}_{\mathbf{k}}$  is Hamiltonian matrix in the standard gauge. In this gauge,  $\mathbf{H}'_{\mathbf{k}}$  is not periodic and obeys the relation  $\mathbf{H}'_{\mathbf{k}+\mathbf{G}} = \mathbf{V}_{\mathbf{G}}^{\dagger} \mathbf{H}'_{\mathbf{k}} \mathbf{V}_{\mathbf{G}}$ , which should be accounted for whenever the eigenstates of the Hamiltonian are needed on the boundary of BZ or outside of it, as occurs in calculations of correlation functions. For this reason we work in the standard gauge here. However, we will also comment on the alternative gauge.

By introducing the relations

$$\hat{c}_{l\mathbf{k}, \sigma} = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_i} \exp(-i\mathbf{k} \cdot \mathbf{R}_i) \hat{c}_{l\mathbf{R}_i, \sigma}, \quad (7)$$

$$\hat{c}_{l\mathbf{R}_i, \sigma} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{R}_i) \hat{c}_{l\mathbf{k}, \sigma}, \quad (8)$$

and substituting them in the Hamiltonian Eq. (4), one obtains

$$\hat{\mathcal{H}} = \sum_{\mathbf{k}, \sigma} \sum_{l_1 l_2} \mathbf{H}_{\mathbf{k}\sigma}^{l_1 l_2} \hat{c}_{l_1 \mathbf{k}, \sigma}^\dagger \hat{c}_{l_2 \mathbf{k}, \sigma}, \quad (9)$$

$$\mathbf{H}_{\mathbf{k}\sigma}^{l_1 l_2} = \sum_{\mathbf{R}_i} e^{-i\mathbf{k} \cdot \mathbf{R}_i} \mathbf{H}_{\mathbf{R}_i, \mathbf{0}}^{l_1 l_2, \sigma},$$

where we used the fact that  $\mathbf{H}_{\mathbf{R}_1, \mathbf{R}_2}^{l_1 l_2, \sigma}$  depends on the relative distance between sites  $\mathbf{R}_1$  and  $\mathbf{R}_2$ , or  $\mathbf{H}_{\mathbf{R}_1 + \mathbf{R}_0, \mathbf{R}_2 + \mathbf{R}_0}^{l_1 l_2, \sigma} = \mathbf{H}_{\mathbf{R}_1, \mathbf{R}_2}^{l_1 l_2, \sigma}$ , and the fact that the ionic potential is periodic  $\sum_{\mathbf{R}_i} V(\mathbf{r} + \mathbf{R}_0 - \mathbf{R}_i) = \sum_{\mathbf{R}_j} V(\mathbf{r} - \mathbf{R}_j)$ . Equation (9) is what we call the Hamiltonian in the orbital basis.

In matrix form, the Hamiltonian in the orbital basis is given by  $\mathcal{H} = \sum_{\mathbf{k}\sigma} \phi_{\mathbf{k}\sigma}^\dagger \mathbf{H}_{\mathbf{k}\sigma} \phi_{\mathbf{k}\sigma}$ , where  $\phi_{\mathbf{k}} \equiv (c_{1, \mathbf{k}\sigma}, \dots, c_{n_{\text{orb}}, \mathbf{k}\sigma})^T$  and  $n_{\text{orb}}$  denotes the number of orbitals within a unit cell (this counts all atoms and their respective orbitals). It is worth recalling that while both  $\phi_{\mathbf{k}\sigma}$  and  $\mathbf{H}_{\mathbf{k}\sigma}$  are basis and gauge dependent, the Hamiltonian  $\mathcal{H}$  is an observable and is gauge invariant.

### III. CHARGE SUSCEPTIBILITY

#### A. Definitions and link between susceptibility of an orbital basis model and observable susceptibility

In this section, we introduce a generalized susceptibility Eq. (23) of the type encountered in orbital-basis calculations and its relation to the susceptibility observable in the continuum, Eq. (22). This section then also defines oscillator matrix elements Eq. (13) and Eq. (17) that are necessary to connect the two.

To be specific, consider the density-density response function given by

$$\chi(\mathbf{r}, \mathbf{r}', \tau) = \langle T_\tau \hat{n}(\mathbf{r}, \tau) \hat{n}(\mathbf{r}', 0) \rangle_c, \quad (10)$$

where the density operator can be written as  $\hat{n}(\mathbf{r}, \tau) = \sum_\sigma \hat{\Psi}_\sigma^\dagger(\mathbf{r}, \tau) \hat{\Psi}_\sigma(\mathbf{r}, \tau)$ . The subscript  $c$  on the average reminds us that we take only the connected part. The operators here evolve in Matsubara imaginary time  $\tau$ . In Fourier space, using the relation  $\chi(\mathbf{r}, \mathbf{r}', \tau) = (1/N) \sum_{\mathbf{R}_0} \chi(\mathbf{r} + \mathbf{R}_0, \mathbf{r}' + \mathbf{R}_0, \tau)$  where  $\mathbf{R}_0$  is a lattice vector, we have

$$\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \tau) = \frac{1}{V} \int d\mathbf{r} d\mathbf{r}' e^{-i(\mathbf{q} + \mathbf{G}) \cdot \mathbf{r}} \chi(\mathbf{r}, \mathbf{r}', \tau) e^{i(\mathbf{q} + \mathbf{G}') \cdot \mathbf{r}'}, \quad (11)$$

where the integral is over the entire volume  $V$ . Here,  $\mathbf{q} + \mathbf{G}'$  and  $\mathbf{q} + \mathbf{G}$  denote respectively the external field and the response wave vectors which may fall outside of the BZ. Similarly by using the field operator expansion Eq. (2) in terms of the atomiclike orbitals we find

$$\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \tau) = \sum_{\sigma\sigma'} \sum_{l_1 \dots l_4} \sum_{\mathbf{R}_1 \dots \mathbf{R}_4} \mathbf{O}_{\mathbf{R}_1 \mathbf{R}_2}^{l_1 l_2, \sigma}(\mathbf{q} + \mathbf{G})$$

$$\times \langle T_\tau \hat{c}_{l_1 \mathbf{R}_1, \sigma}^\dagger(\tau) \hat{c}_{l_2 \mathbf{R}_2, \sigma}(\tau) \hat{c}_{l_3 \mathbf{R}_3, \sigma'}^\dagger(0) \hat{c}_{l_4 \mathbf{R}_4, \sigma'}(0) \rangle_c$$

$$\times \mathbf{O}_{\mathbf{R}_3 \mathbf{R}_4}^{l_3 l_4, \sigma'}(-\mathbf{q} - \mathbf{G}'), \quad (12)$$

where we defined the oscillator matrix element as

$$\mathbf{O}_{\mathbf{R}_1 \mathbf{R}_2}^{l_1 l_2, \sigma}(\mathbf{q}) \equiv \frac{1}{\sqrt{V}} \int d\mathbf{r} e^{-i\mathbf{q} \cdot \mathbf{r}} \phi_{\mathbf{R}_1 l_1, \sigma}^*(\mathbf{r}) \phi_{\mathbf{R}_2 l_2, \sigma}(\mathbf{r}). \quad (13)$$

Using the relation,  $\phi_{\mathbf{R}_1 + \mathbf{R}_0, l_1, \sigma}(\mathbf{r}) = \phi_{\mathbf{R}_1, l_1, \sigma}(\mathbf{r} - \mathbf{R}_0)$ , one finds that the oscillator matrix elements satisfy

$$\mathbf{O}_{\mathbf{R}_1 + \mathbf{R}_0, \mathbf{R}_2 + \mathbf{R}_0}^{l_1 l_2, \sigma}(\mathbf{q}) = \exp(-i\mathbf{q} \cdot \mathbf{R}_0) \mathbf{O}_{\mathbf{R}_1, \mathbf{R}_2}^{l_1 l_2, \sigma}(\mathbf{q}). \quad (14)$$

Substituting the Fourier-space basis for the second-quantized operators, Eq. (8), in Eq. (12) for the susceptibility, one finds

$$\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q}, \tau) = \sum_{\sigma\sigma'} \sum_{\mathbf{k}\mathbf{k}'} \sum_{l_1 \dots l_4} \mathbf{O}_{\mathbf{k}, \mathbf{k} + \mathbf{q}}^{l_1 l_2, \sigma}(\mathbf{q} + \mathbf{G})$$

$$\times \langle T_\tau \hat{c}_{\mathbf{k} l_1, \sigma}^\dagger(\tau) \hat{c}_{\mathbf{k} + \mathbf{q} l_2, \sigma}(\tau) \hat{c}_{\mathbf{k}' l_3, \sigma}^\dagger(0) \hat{c}_{\mathbf{k}' - \mathbf{q} l_4, \sigma}(0) \rangle_c$$

$$\times \mathbf{O}_{\mathbf{k}', \mathbf{k}' - \mathbf{q}}^{l_3 l_4, \sigma'}(-\mathbf{q} - \mathbf{G}'), \quad (15)$$

where it is understood that  $\mathbf{k} + \mathbf{q}$  and  $\mathbf{k}' - \mathbf{q}$  must be folded back to the first Brillouin zone with the appropriate Umklapp reciprocal lattice vectors  $\mathbf{G}_0$  and  $\mathbf{G}'_0$ , as follows from the following identity:

$$\sum_{\mathbf{R}_1 \mathbf{R}_2} e^{-i\mathbf{k}_1 \cdot \mathbf{R}_1} e^{i\mathbf{k}_2 \cdot \mathbf{R}_2} \mathbf{O}_{\mathbf{R}_1 \mathbf{R}_2}^{l_1 l_2, \sigma}(\mathbf{q})$$

$$= \frac{1}{N} \sum_{\mathbf{R}_1 \mathbf{R}_2 \mathbf{R}_0} e^{i\mathbf{q} \cdot \mathbf{R}_0} e^{-i\mathbf{k}_1 \cdot \mathbf{R}_1} e^{i\mathbf{k}_2 \cdot \mathbf{R}_2} \mathbf{O}_{\mathbf{R}_1 + \mathbf{R}_0, \mathbf{R}_2 + \mathbf{R}_0}^{l_1 l_2, \sigma}(\mathbf{q})$$

$$= \delta_{\mathbf{k}_1, \mathbf{k}_2 - \mathbf{q} + \mathbf{G}_0} \mathbf{O}_{\mathbf{k}_1 \mathbf{k}_2}^{l_1 l_2, \sigma}(\mathbf{q}), \quad (16)$$

with

$$\mathbf{O}_{\mathbf{k}_1 \mathbf{k}_2}^{l_1 l_2, \sigma}(\mathbf{q}) = \frac{1}{N} \sum_{\mathbf{R}_1 \mathbf{R}_2} e^{-i\mathbf{k}_1 \cdot \mathbf{R}_1} \mathbf{O}_{\mathbf{R}_1 \mathbf{R}_2}^{l_1 l_2, \sigma}(\mathbf{q}) e^{i\mathbf{k}_2 \cdot \mathbf{R}_2}$$

$$= \frac{1}{\sqrt{V}} \int d\mathbf{r} e^{-i\mathbf{q} \cdot \mathbf{r}} \phi_{l_1 \mathbf{k}_1}^*(\mathbf{r}) \phi_{l_2 \mathbf{k}_2}(\mathbf{r}). \quad (17)$$

We thus obtained the expression for the response function in an orbital basis that can be used for model calculations.

For a noninteracting system, the Bloch function basis set is usually preferred. It is useful to see how the oscillator matrix elements come in this representation. The time evolution of the operators in Eq. (15) in this case are trivial and one can evaluate the four-point correlation function to obtain the following expression for the response function [SU(2) symmetric case] in Matsubara-frequency space

$$\chi_{\mathbf{G}\mathbf{G}'}^{0, ph}(\mathbf{q}, \nu_n) = -\frac{1}{N} \sum_{\mathbf{k}\sigma} \sum_{n\mathbf{m}} \frac{f(\epsilon_{\mathbf{m}, \mathbf{k} - \mathbf{q}}) - f(\epsilon_{\mathbf{n}, \mathbf{k}})}{\hbar \nu_n + \epsilon_{\mathbf{m}, \mathbf{k} - \mathbf{q}} - \epsilon_{\mathbf{n}, \mathbf{k}}} \langle \psi_{n\mathbf{k} - \mathbf{q}} |$$

$$\times e^{-i(\mathbf{q} + \mathbf{G}) \cdot \hat{\mathbf{r}}} | \psi_{m\mathbf{k}} \rangle \langle \psi_{m\mathbf{k}} | e^{i(\mathbf{q} + \mathbf{G}') \cdot \hat{\mathbf{r}}} | \psi_{n\mathbf{k} - \mathbf{q}} \rangle, \quad (18)$$

where  $\hat{\mathbf{r}}$  denotes the position operator,  $\psi_{n\mathbf{k}}$  are the Bloch states, and  $\chi_{\mathbf{G}\mathbf{G}'}^{0, ph}(\mathbf{q}, \nu_n) \equiv \chi^{0, ph}(\mathbf{q} + \mathbf{G}, \mathbf{q} + \mathbf{G}', \nu_n)$ . Hereafter, we drop the reciprocal lattice vectors. They can easily be added when necessary.

For a weakly to moderately correlated system it is useful to begin with Eq. (18) and use the perturbation expansion to consider correlation effects. In this approach, Eq. (18) should be rewritten in the orbital basis which is the natural basis for including interaction effects. Expanding the Bloch functions in an atomiclike orbital basis set, namely,

$$\psi_{n\mathbf{k}}(\mathbf{r}) = \sum_l a_{l\mathbf{k}}^n \phi_{l\mathbf{k}}(\mathbf{r}), \quad (19)$$

with the definition  $a_{nk}^l = \langle \phi_{lk} | \psi_{nk} \rangle$ , the matrix element for charge susceptibility can be rewritten as

$$\langle \psi_{nk-\mathbf{q}} | e^{-i\mathbf{q}\cdot\mathbf{r}} | \psi_{mk} \rangle = \sum_{l_1 l_2} a_{nk-\mathbf{q}}^{l_1*} a_{mk}^{l_2} \mathbf{O}_{\mathbf{k}}^{l_1 l_2}(\mathbf{q}), \quad (20)$$

where the oscillator matrix elements in the orbital basis are given by Eq. (17) with the shorthand [SU(2) symmetric case]

$$\mathbf{O}_{\mathbf{k}}^{l_1 l_2}(\mathbf{q}) = \mathbf{O}_{\mathbf{k}-\mathbf{q}, \mathbf{k}}^{l_1 l_2, \sigma}(\mathbf{q}). \quad (21)$$

Using this expression in the response function, one finds

$$\begin{aligned} \chi_{ph}^0(\mathbf{q}, \nu_n) &= \frac{1}{N} \sum_{\mathbf{k}\sigma} \sum_{l_1 l_2, l_3 l_4} [\chi_{ph}^0(\mathbf{q}, \nu_n)]_{\mathbf{k}}^{l_1 l_2, l_3 l_4} \\ &\quad \times \mathbf{O}_{\mathbf{k}}^{l_1 l_2}(\mathbf{q}) \mathbf{O}_{\mathbf{k}}^{l_3 l_4}(-\mathbf{q}), \end{aligned} \quad (22)$$

where we defined the *generalized susceptibility* by

$$\begin{aligned} [\chi_{ph}^0(\mathbf{q}, i\nu_n)]_{\mathbf{k}}^{l_1 l_2, l_3 l_4} &\equiv - \sum_{nm} \frac{f(\epsilon_{m, \mathbf{k}-\mathbf{q}}) - f(\epsilon_{n, \mathbf{k}})}{i\nu_n + \epsilon_{m, \mathbf{k}-\mathbf{q}} - \epsilon_{n, \mathbf{k}}} \\ &\quad \times a_{nk-\mathbf{q}}^{l_1*} a_{mk}^{l_2} a_{mk}^{l_3*} a_{nk-\mathbf{q}}^{l_4}. \end{aligned} \quad (23)$$

It describes the independent propagation of a p-h excitation and is essential to study charge and magnetic response functions in interacting systems (see below). It appears in the response to an external field and it solely depends on the electronic structure. This is the quantity that is usually computed and analyzed in the literature, instead of the response function Eq. (22). In other words, the oscillator matrix elements are usually neglected. Note that when orbital indices are not explicitly written, generalized susceptibilities are identified by a bold symbol to emphasize the tensor character. The same symbol but without boldface is used for the corresponding observable susceptibilities.

The generalized susceptibility of an interacting system is called the dressed generalized susceptibility,  $\chi_{ph}(\mathcal{Q})$ . It is different from  $\chi_{ph}^0(\mathcal{Q})$  because propagating p-h pairs interact with their environment and with each other. In a perturbation approach, the dressed generalized susceptibility can be decomposed into bare susceptibility,  $\chi_{ph}^0(\mathcal{Q})$ , and vertex corrections,  $\mathbf{X}_{ph}(\mathcal{Q})$ , where we defined  $\mathcal{Q} \equiv (\mathbf{q}, \nu_n)$  as the momentum-frequency four-vectors.

With the help of the irreducible vertex functions in the density channel,  $\Gamma^{\text{irr}, d}(\mathcal{Q}) = \Gamma^{\text{irr}, \uparrow\downarrow}(\mathcal{Q}) + \Gamma^{\text{irr}, \uparrow\uparrow}(\mathcal{Q})$ , the equation for the generalized dressed susceptibility in this channel is [12,13]

$$\begin{aligned} \chi_{ph}^d(\mathcal{Q}) &= \chi_{ph}^0(\mathcal{Q}) + \mathbf{X}_{ph}^d(\mathcal{Q}) \\ &= \chi_{ph}^0(\mathcal{Q}) - \chi_{ph}^d(\mathcal{Q}) \Gamma^{\text{irr}, d}(\mathcal{Q}) \chi_{ph}^0(\mathcal{Q}), \end{aligned} \quad (24)$$

which can be rewritten as

$$\chi_{ph}^d(\mathcal{Q}) = \chi_{ph}^0(\mathcal{Q}) [\mathbf{1} + \Gamma^{\text{irr}, d}(\mathcal{Q}) \chi_{ph}^0(\mathcal{Q})]^{-1}. \quad (25)$$

Note that at the above two equations a summation over all internal wave vectors, frequencies, and orbital indices is assumed. The generalized susceptibilities are tensor quantities.

In order to obtain the observable charge response, the external legs of the generalized susceptibility  $\chi_{ph}^d(\mathcal{Q})$  should

be closed, just as the noninteracting case, Eq. (22), by the oscillator matrix elements, Eq. (17). Therefore, the observable dressed correlation function is given by an equation similar to Eq. (22) where  $\chi_{ph}^0(\mathbf{q}, \nu_n)$  is replaced by  $\chi_{ph}^d(\mathbf{q}, \nu_n)$ . Furthermore, as discussed in Appendix A, the oscillator matrix elements also appear in the interaction matrix elements. While for local interactions one can absorb these matrix elements in the definition of the interaction, further-neighbor interactions necessitate extra phase factors that come from the oscillator matrix elements.

Inspecting the observable response function of the system in the normal phase provides the means to characterize a phase transition and identify the responsible driving mechanism. The response function becomes singular at a continuous (second order) phase transition into an ordered phase. Since the oscillator matrix elements are temperature independent and nonsingular, the singularity of the observable dressed correlation function coincides with the singularity of the  $\chi_{ph}^d(\mathbf{q}, \nu_n)$ . From Eq. (25) one can see that  $\chi_{ph}^d(\mathbf{q}, \nu_n)$  becomes singular once  $-\Gamma^{\text{irr}, d}(\mathbf{q}) \chi_{ph}^0(\mathbf{q}, i\nu_n = 0)$  has an eigenvalue equal to one. Hence the distance from a density transition is commonly measured by the largest (dimensionless) eigenvalue of  $-\Gamma^{\text{irr}, d}(\mathbf{q}) \chi_{ph}^0(\mathbf{q}, i\nu_n = 0)$ . This is the so-called density Stoner factor,  $\alpha_{\mathbf{q}}^d$ . An instability occurs once the Stoner factor becomes unity. The corresponding wave vector is the wave vector of the instability. The Stoner factor analysis describes the tendency of the ground state towards a density or magnetic instability, independent of the external field.

## B. Random phase approximation

As one can see from Eq. (22), the connection between theoretical and experimental results requires a calculation of the oscillator matrix elements. The equation for the oscillator matrix elements, Eq. (13), can be rewritten as

$$\begin{aligned} \mathbf{O}_{\mathbf{R}_1 \mathbf{R}_2}^{l_1 l_2, \sigma}(\mathbf{q}) &= \exp[-i\mathbf{q} \cdot (\mathbf{R}_1 + \mathbf{r}_{\alpha_1} + \delta/2)] \frac{1}{\sqrt{V}} \int d\mathbf{r} e^{-i\mathbf{q}\cdot\mathbf{r}} \\ &\quad \times \phi_{l_1, \sigma}^*(\mathbf{r} + \delta/2) \phi_{l_2, \sigma}(\mathbf{r} - \delta/2), \end{aligned} \quad (26)$$

where  $\delta \equiv (\mathbf{R}_2 + \mathbf{r}_{\alpha_2}) - (\mathbf{R}_1 + \mathbf{r}_{\alpha_1})$ . Note that the second line at the above equation *only* depends on the spatial distance of the orbital centers and is translationally invariant. It is clear that one cannot get rid of the phase in the prefactor by changing the gauge since that phase depends on the center of mass coordinate.

In practice, a commonly used approximation is the so-called random phase approximation (RPA). In this approximation, one assumes a static irreducible vertex, and only oscillator matrix elements with identical orbital centers and character are kept, i.e.,

$$\mathbf{O}_{\mathbf{R}_1 \mathbf{R}_2}^{l_1 l_2, \sigma}(\mathbf{q}) \propto \exp[-i\mathbf{q} \cdot (\mathbf{R}_1 + \mathbf{r}_{\alpha})] \delta_{l_1 l_2} \delta_{\mathbf{R}_1, \mathbf{R}_2}, \quad (27)$$

where orbital  $l_1$  belongs to atom  $\alpha$ . This approximation is only justified in the small wave vector  $\mathbf{q} \rightarrow \mathbf{0}$  or very large wavelength limit where the inner structure of the unit cell is irrelevant. Actually, the oscillator matrix elements with nearest-neighbor  $l_1$  and  $l_2$  are not negligible for finite  $\mathbf{q}$ . Indeed, the hopping matrix elements are given by a similar overlap

integral where the phase factor is replaced by the Laplacian, see Eq. (4), which oscillates much faster.

Within this approximation, the oscillator matrix element in Fourier space Eq. (17) is proportional to

$$\mathbf{O}_{\mathbf{k}}^{l_1 l_2, \sigma}(\mathbf{q}) \propto \exp(-i\mathbf{q} \cdot \mathbf{r}_\alpha) \delta_{l_1 l_2}, \quad (28)$$

which allows one to perform the  $\mathbf{k}$  summation in Eq. (22). Hence the resulting observable response function is given by

$$\chi_{\text{ph}}^0(\mathbf{q}, \nu_n) = \sum_{l_1 l_2} [\chi_{\text{ph}}^{0, \text{RPA}}(\mathbf{q}, \nu_n)]_{l_1 l_1, l_2 l_2} \exp[-i\mathbf{q} \cdot (\mathbf{r}_\alpha - \mathbf{r}_\beta)], \quad (29)$$

where  $l_1$  and  $l_2$  orbitals belong to atoms  $\alpha$  and  $\beta$ , respectively. In many studies the phase factor in Eq. (29) is incorrectly neglected. The so-called irreducible RPA generalized susceptibility in the above equation can be rewritten in terms of the Green's function as

$$[\chi_{\text{ph}}^{0, \text{RPA}}(Q)]_{l_1 l_1, l_2 l_2} = -\left(\frac{k_B T}{N}\right) \sum_K \mathbf{G}_{K-Q, l_1 l_2} \mathbf{G}_{K, l_2 l_1}, \quad (30)$$

where  $K \equiv (\mathbf{k}, \omega_m)$ . It is worth emphasizing that Eq. (27), Eq. (28), and Eq. (29) are gauge independent. The difference between systems with symmorphic space-group symmetry and those with nonsymmorphic space-group symmetry appears here. For systems with symmorphic space group, the generalized susceptibility has the same symmetries as the response function, while for systems with nonsymmorphic space group it is necessary to take into account the oscillator matrix elements to obtain the observed symmetry of the response function.

The correct observable symmetries are also recovered in the RPA approximation for the dressed observable susceptibility. Closing the external legs of the dressed generalized susceptibility by Eq. (28) yields an equation similar to Eq. (29) where  $\chi_{\text{ph}}^0(\mathbf{q}, \nu_n)$  is replaced by  $\chi_{\text{ph}}^d(\mathbf{q}, \nu_n)$ . The lowest correction to the bare observable correlation function is

$$-\sum_{l_1 \dots l_6} e^{-i\mathbf{q} \cdot \mathbf{r}_\alpha} [\chi_{\text{ph}}^{0, \text{RPA}}(\mathbf{q}, \nu_n)]_{l_1 l_1, l_3 l_4} \times [\Gamma_{\text{RPA}}^{d, \text{irr}}(\mathbf{q}, \nu_n)]_{l_3 l_4, l_5 l_6} [\chi_{\text{ph}}^{0, \text{RPA}}(\mathbf{q}, \nu_n)]_{l_5 l_6, l_2 l_2} e^{i\mathbf{q} \cdot \mathbf{r}_\beta}, \quad (31)$$

where  $l_1$  and  $l_2$  orbitals belong to atoms  $\alpha$  and  $\beta$ , respectively, and  $\Gamma_{\text{RPA}}^{d, \text{irr}}$  denotes the irreducible vertex function in the RPA approximation. In the case of a local interaction,  $l_3 \dots l_6$  orbitals belong to the same atom  $\gamma$ . By multiplying the above equation with  $\exp[-i\mathbf{q} \cdot (\mathbf{r}_\gamma - \mathbf{r}_\gamma)]$  one can show that the resulting expression has the same symmetry as the observable bare susceptibility and therefore the interacting correlation function has it as well. Extension to the case of nonlocal interaction is straightforward. As discussed in Appendix A, nonlocal interaction matrix elements acquire extra phase factors that come from the oscillator matrix elements.

#### IV. NONSYMMORPHIC SYMMETRY OPERATIONS AND HAMILTONIAN TRANSFORMATION

In this section we discuss how the basis dependent  $\mathbf{H}_{\mathbf{k}}$ , Eq. (9), transforms under point group symmetry operations

of the lattice. This is a necessary step to demonstrate how oscillator matrix elements allow response functions to exhibit, at certain wave vectors, symmetries that are absent from  $\mathbf{H}_{\mathbf{k}}$ . We restrict ourselves to the interesting case of the iron-pnictide superconductors with a nonsymmorphic point group. With 16 Fe- $d$  and As- $p$  orbitals in the unit cell, the Hamiltonian in the orbital basis is given by  $\mathcal{H} = \sum_{\mathbf{k}} \phi_{\mathbf{k}}^\dagger \mathbf{H}_{\mathbf{k}} \phi_{\mathbf{k}}$ , where  $\phi_{\mathbf{k}} \equiv (c_{1, \mathbf{k}}, \dots, c_{16, \mathbf{k}})^T$ .

In general, a nontranslation member of the space group can be described by  $\{g|\boldsymbol{\tau}\}$ , where  $g$  is a point group operation (rotation, reflection, etc.), which keeps at least one point (the coordinate origin) invariant, followed by a *half-integer* translation by vector  $\boldsymbol{\tau}$  [6]. The half-integer translation vectors depend on the choice of the origin and axis. They cannot be eliminated by shifting the origin for screw axis and glide-plane operations. A space group including these symmetries is called nonsymmorphic. Figure 1 sketches the lattice structure of a trilayer Fe-As unit and three generators of the  $P4/nmm$  group (see Appendix B for a full table of the symmetry operations). The origin is chosen to be on the inversion center in the middle of an Fe-Fe link. We consider the three generators  $\{C_{2y}|0\frac{1}{2}0\}$ ,  $\{C_{2a}|000\}$ , and inversion  $\{I|000\}$ , where  $C_{nr}$  is a counterclockwise rotation through  $2\pi/n$  radians about the axis labeled by  $\mathbf{r}$ . The  $\{C_{2y}|0\frac{1}{2}0\}$  operation is a screw axis operation where a rotation is followed by a translation along the rotation axis. For a nonsymmorphic space group, the nontranslation elements  $\{g|\boldsymbol{\tau}\}$  do not form a group, since their combination may turn out to be a pure lattice translation. However, the symmorphic elements  $\{g|\mathbf{0}\}$  by themselves form a group.

Consider an atom  $\alpha$  at the  $i$ th unit cell located at  $\mathbf{R}_i + \mathbf{r}_\alpha$ . Under a symmetry operation,  $\{g|\boldsymbol{\tau}\}$ , this atom transfers to  $g\mathbf{R}_i + g\mathbf{r}_\alpha + \boldsymbol{\tau}$ . This is accompanied by a rotation in the associated orbital space along with a possible atom index exchange  $\alpha \rightarrow \alpha'$  if there is more than one atom of the same kind in the unit cell. The peculiarity of a nonsymmorphic operation is that the new position may be in a unit cell adjacent to  $g\mathbf{R}_i$ . Defining  $\mathbf{L}_\alpha$  as a real space lattice vector which connects atoms  $\alpha'$  in these two unit cells, we have  $g\mathbf{r}_\alpha + \boldsymbol{\tau} = \mathbf{r}_{\alpha'} + \mathbf{L}_\alpha$ . The vector  $\mathbf{L}_\alpha$  depends on the position of atom  $\alpha$  and on the vector  $\boldsymbol{\tau}$ . For example, under  $\{C_{2y}|0\frac{1}{2}0\}$  the  $B$  site of the unit cell at origin transfers to the  $A$  site of the neighboring unit cell in the  $y$  direction, while the  $A$  site transfers to the  $B$  site of the same unit cell (see Fig. 1). Thus, for this symmetry operation,  $\mathbf{L}_\alpha = \mathbf{0}$  for sublattice  $A$  and  $\mathbf{L}_\alpha = \mathbf{a}_2$  for sublattice  $B$ , where  $\mathbf{a}_j$  denotes the real-space lattice unit vector in  $j$ th direction. In Fourier space, these vectors  $\mathbf{L}_\alpha$  introduce atom-dependent extra phases.

As shown in Appendix C, under a symmetry operation,  $\{g|\boldsymbol{\tau}\}$ , the electron annihilation operators in the orbital basis  $\phi_{\mathbf{k}}$  transform as  $\{g|\boldsymbol{\tau}\}\phi_{\mathbf{k}} = \mathbf{U}'_{g\mathbf{k}, \mathbf{L}} \mathbf{U}_g \phi_{g\mathbf{k}}$ , where  $\mathbf{U}_g$  is a  $16 \times 16$  unitary matrix that describes the effect of the  $\{g|\boldsymbol{\tau}\}$  operation in the orbital basis and where the action of  $g$  on  $\mathbf{k}$  is represented by a  $3 \times 3$  orthogonal matrix. The existence of the above mentioned  $\mathbf{L}_\alpha$  vectors leads to a diagonal  $16 \times 16$  unitary matrix  $\mathbf{U}'_{g\mathbf{k}, \mathbf{L}}$  with element  $\exp(-ig\mathbf{k} \cdot \mathbf{L}_\alpha)$ . Therefore, the invariance of the Hamiltonian takes the form

$$\mathbf{H}_{g\mathbf{k}} = \mathbf{U}'_g \mathbf{U}'_{g\mathbf{k}, \mathbf{L}} \mathbf{H}_{\mathbf{k}} \mathbf{U}'_{g\mathbf{k}, \mathbf{L}} \mathbf{U}_g, \quad (32)$$

which is different from the result for a symmorphic operation, i.e.,

$$\mathbf{H}_{g\mathbf{k}} = \mathbf{U}_g^\dagger \mathbf{H}_{\mathbf{k}} \mathbf{U}_g. \quad (33)$$

For iron-based superconductors, the usual relation, Eq. (33), is recovered under the  $\{I|000\}$  and  $\{C_{2a}|000\}$  generators where the extra phase factors are absent. However, under  $\{C_{2y}|0\frac{1}{2}0\}$  only one sublattice obtains the extra phase,  $\exp[-i2\pi(g\mathbf{k})_y]$ , where  $(g\mathbf{k})_y$  is the  $y$  component of the wave vector in reciprocal lattice vector units. This implies that for this operation Eq. (33) is valid only for the intrasublattice part of the Hamiltonian, where the extra phases are canceled out, and not for the whole matrix. The same is true for all the symmetry operations of the group with nonzero  $\tau$ . It is not possible to eliminate these phases with a gauge transformation for the same reason that it is not possible to eliminate the half-integer translation with a shift of origin. These symmetry properties are clarified further using a minimal model Hamiltonian in Appendixes D and E.

Given the above symmetry properties for  $\mathbf{H}_{\mathbf{k}}$ , a key question is how the observables recover the full symmetry of the lattice in the absence of any symmetry breaking phase transition. Band energies and their degeneracy are clearly invariant under all symmetry operations. This can be seen from the invariance of  $\det(\omega\mathbf{1} - \mathbf{H}_{\mathbf{k}})$  under symmetry operations and the fact that the determinant of the matrix product is equal to the product of the determinants. Another important quantity, connected to electron charge density, is the electron momentum distribution function, which is given by  $n(\mathbf{k}) = 1 + 2 \operatorname{Re} \sum_{\omega_m} \operatorname{Tr}[\mathbf{O}_{\mathbf{k}}(\mathbf{q} = \mathbf{0})\mathbf{G}(\mathbf{k}, i\omega_m)]$ . Here,  $\mathbf{G}(\mathbf{k}, i\omega_m) = [(i\omega_m + \mu)\mathbf{1} - \mathbf{H}_{\mathbf{k}}]^{-1}$  denotes the electron propagator. Since  $\mathbf{O}_{\mathbf{k}}^{l_1 l_2}(\mathbf{q} = \mathbf{0}) \propto \delta_{l_1 l_2}$  as can be seen from Eq. (17), only intraorbital components contribute in the trace; hence this quantity is also invariant, in the usual sense, under all symmetry operations. The same is true for diagonal components of the spectral function.

Coming back to the susceptibility, both the generalized susceptibility and the observable ones behave the same way under a symmorphic point-group operation. Such a transformation permutes atomic orbitals into one another and changes positions following  $\mathbf{r}_\alpha \rightarrow g\mathbf{r}_\alpha$ . In Fourier components this translates into  $\mathbf{q} \rightarrow g^{-1}\mathbf{q}$  and the phase factor coming from the oscillator matrix elements in the calculation of the observable susceptibility Eq. (29) has the same property.

For a nonsymmorphic operation on the generalized susceptibility, there is an additional phase  $\exp[-i2\pi(g\mathbf{k})_y]$ , where  $(g\mathbf{k})_y$  is the  $y$  component of the wave vector in reciprocal lattice vector units, that depends on the position of the atom in the unit cell. The phase factor in the transformation from the generalized susceptibility to the observable susceptibility Eq. (29) has an analogous dependence on position within the unit cell that compensates that phase and  $C_{4z}$  symmetry at  $q_z = 0$  is recovered.

In the following section we illustrate further the more subtle case of the symmetry properties of the generalized susceptibilities by performing explicit calculations for LiFeAs. We show that the generalized susceptibilities do not have  $C_{4z}$  symmetry at  $q_z = 0$ . The full symmetries are recovered only when the generalized susceptibilities are embedded back in the continuum using appropriate matrix elements between basis states, as discussed above. In Appendixes D and E we perform

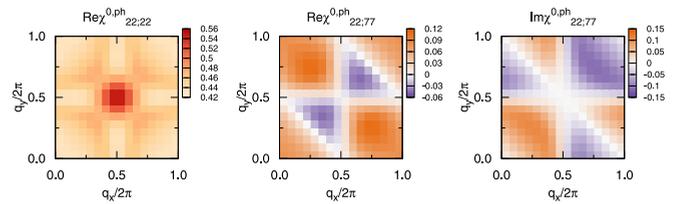


FIG. 2. Real and imaginary parts of the  $d_{x^2-y^2}$  intraorbital, intrasublattice 22;22 (left), and intersublattice 22;77 (middle and right) component of the in-plane bare generalized susceptibility at the lowest bosonic frequency,  $[\chi_{\text{ph}}^{0,\text{RPA}}(\mathbf{q}, \nu_n = 0)]$ , of LiFeAs at  $k_B T = 0.01$  eV in the p-h channel. The Kohn-Sham eigenstates are projected to atomlike Wannier basis to construct the Hamiltonian in the orbital basis [15]. For  $K$  summation in Eq. (23) we have used a  $32 \times 32 \times 16$   $k$ -mesh and 1024 positive frequencies.

the full calculation for a minimal tight-binding Hamiltonian to illustrate in detail what happens.

## V. GENERALIZED SUSCEPTIBILITY OF LiFeAs

Here, we present some results for the in-plane generalized susceptibilities in the RPA approximation. Although our discussion is general, to be specific we present results on LiFeAs [14]. We are particularly interested in fourfold symmetry in the normal state. We show explicitly that nonsymmorphic operations lead to generalized susceptibilities that do not have  $C_{4z}$  symmetry even at  $q_z = 0$ . This symmetry is recovered in observable susceptibilities if we take into account oscillator matrix elements.

We work in the two-Fe unit cell and all Fe-3d and As-4p orbitals are considered in the calculations. In what follows, we focus on the  $d$  orbitals of Fe-1 (Fe-2) (on  $A$  and  $B$  sublattices, respectively) that have large weight around the Fermi level:  $d_{x^2-y^2}$  will be referred to as 2(7) and  $d_{xz}$  and  $d_{yz}$  orbitals as 4(9) and 5(10). Note that in the one-Fe unit cell with a coordinate system rotated by  $\pi/4$  around the  $z$  axis relative to the two-Fe coordinate system, the  $d_{x^2-y^2}$  orbital becomes  $d_{xy}$ .

The left panel of Fig. 2 shows the in-plane bare generalized susceptibility  $[\chi_{\text{ph}}^{0,\text{RPA}}]_{22;22} (= [\chi_{\text{ph}}^{0,\text{RPA}}]_{77;77})$  of LiFeAs at the lowest bosonic Matsubara frequency. It is the dominant component of the bare susceptibility with incommensurate peaks around the  $M$  point, which comes from nesting between the hole and electron pockets. [We use the term incommensurate in a general sense to distinguish from high symmetry points such as the  $M$  point. With this definition, a wave vector  $(\pi/3, \pi)$  is incommensurate.] The  $d_{xz(yz)}$  intraorbital components,  $[\chi_{\text{ph}}^{0,\text{RPA}}]_{44;44}$  and  $[\chi_{\text{ph}}^{0,\text{RPA}}]_{55;55}$ , show commensurate peaks at the  $M$  point (not shown) [13,16]. These components are obtained from the convolution of two intrasublattice propagators and therefore they are invariant under all symmetry operations. The  $d_{x^2-y^2}$  component has fourfold symmetry while the  $d_{xz(yz)}$  components are degenerate and are transformed into each other under fourfold operations.

However, a general component of the in-plane bare generalized susceptibility does not have fourfold symmetry because the corresponding symmetry operator is nonsymmorphic. For example, the middle and right panels of Fig. 2 show the real and imaginary parts of the in-plane  $[\chi_{\text{ph}}^{0,\text{RPA}}]_{22;77}$

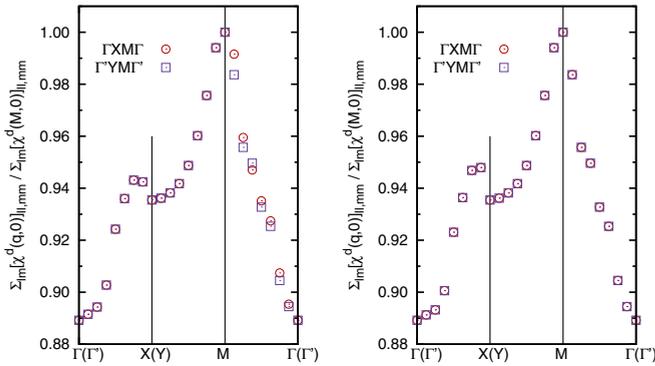


FIG. 3. RPA observable dressed susceptibility in density channel of LiFeAs at  $k_B T = 0.01$  eV, without (left) and with (right) the oscillator matrix elements [see Eq. (29)]. The fourfold symmetry is absent for wave vectors within the BZ (compare  $\Gamma - M$  and  $\Gamma' - M$  paths on the left panel) if one neglects the phases due to the oscillator matrix elements (left panel). This symmetry is recovered by including these phases (right panel). The  $\Gamma - M$  path is along the primary diagonal (positive slope), while the  $\Gamma' - M$  path is along the secondary diagonal (negative slope). The screened interaction parameters used in the RPA dressing are  $J_s = 0.1U_s$  and  $U_s = 1.32$  eV [13]. The interorbital interaction and pair hopping are determined assuming spin-rotational symmetry.

component as an example of components that include inter-sublattice propagators. Since  $\mathbf{G}_{27}(K + Q)$  and  $\mathbf{G}_{72}(K)$  in the susceptibility involve different wave vectors and Fe sites, the nonsymmorphic phases do not cancel. These components have a lower symmetry than those including only intrasublattice propagators. This leads to a generalized susceptibility without fourfold symmetry.

Figure 3 demonstrates  $\sum_{l,m} [\chi_{\text{ph}}^0(\mathbf{q})]_{ll,mm}$  normalized by  $\sum_{l,m} [\chi_{\text{ph}}^0(M)]_{ll,mm}$  along a high symmetry path (left panel). Despite the fact that the bare charge susceptibility along the main axes shows  $C_4$  symmetry the susceptibility along the primary  $\Gamma - M$  and secondary  $\Gamma' - M$  diagonals are slightly different. However, taking into account the role played by oscillator matrix elements through Eq. (29) leads to a response function which has fourfold symmetry (right panel).

Finally, in spin-fluctuation mediated pairing, the density/magnetic fluctuations contribute to the pairing interaction. These fluctuations are expressed in terms of the dressed generalized susceptibilities and vertex functions in density and magnetic channels [13,16]. Hence the anisotropy of the generalized susceptibility would influence the superconducting phase as well. Solving the Eliashberg equation yields the leading gap functions which either preserve the full symmetry or have a lower symmetry. Imposing an artificial fourfold symmetry on the generalized susceptibilities, as is done in one-iron unit cell studies, could impact the competition between the leading gap functions and could switch their order.

## VI. CONCLUSION

In conclusion, we clarified the effects of the nonsymmorphic symmetry operations on the Hamiltonian and on the response and correlation functions in the orbital basis. These symmetry operations include half-integer translations that

introduce extra phases when applied to the Hamiltonian. As an example, the fourfold rotational symmetry of the iron-based pnictide LiFeAs that is expected for  $q_z = 0$  does not exist for the nondiagonal part of the Hamiltonian. We showed that, while single-particle eigenvalues maintain full fourfold symmetry, symmetry is only recovered for observable response functions when one takes into account the symmetry properties of both the generalized susceptibility and of the oscillator matrix elements. This was further clarified using a minimal tight-binding Hamiltonian for iron pnictides, which have a nonsymmorphic space group.

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## APPENDIX A: INTERACTION MATRIX ELEMENTS

The interaction term is given by

$$H_{\text{int}} = \frac{1}{2} \sum_{\sigma\sigma'} \frac{1}{V} \int d\mathbf{r} d\mathbf{r}' \Psi_{\sigma}^{\dagger}(\mathbf{r}) \Psi_{\sigma'}^{\dagger}(\mathbf{r}') \times \mathcal{V}(\mathbf{r} - \mathbf{r}') \Psi_{\sigma'}(\mathbf{r}') \Psi_{\sigma}(\mathbf{r}). \quad (\text{A1})$$

Using the field operator expansion in the orbital basis, the interaction term can be rewritten as

$$H_{\text{int}} = \frac{1}{2} \sum_{\sigma\sigma'} \sum_{l_1 \dots l_4} \sum_{\mathbf{R}_1 \dots \mathbf{R}_4} \mathcal{V}_{\mathbf{R}_1 \mathbf{R}_2; \mathbf{R}_3 \mathbf{R}_4}^{l_1, l_2; l_3, l_4} \times \hat{c}_{\mathbf{R}_1 l_1, \sigma}^{\dagger} \hat{c}_{\mathbf{R}_2 l_2, \sigma'}^{\dagger} \hat{c}_{\mathbf{R}_3 l_3, \sigma'} \hat{c}_{\mathbf{R}_4 l_4, \sigma}, \quad (\text{A2})$$

with

$$\mathcal{V}_{\mathbf{R}_1 \mathbf{R}_2; \mathbf{R}_3 \mathbf{R}_4}^{l_1, l_2; l_3, l_4} \equiv \frac{1}{V} \int d\mathbf{r} d\mathbf{r}' \phi_{\mathbf{R}_1 l_1}^*(\mathbf{r}) \phi_{\mathbf{R}_2 l_2}^*(\mathbf{r}') \times \mathcal{V}(\mathbf{r} - \mathbf{r}') \phi_{\mathbf{R}_3 l_3}(\mathbf{r}') \phi_{\mathbf{R}_4 l_4}(\mathbf{r}). \quad (\text{A3})$$

One can easily see that  $\mathcal{V}_{\mathbf{R}_1 \mathbf{R}_2; \mathbf{R}_3 \mathbf{R}_4}^{l_1, l_2; l_3, l_4} = \mathcal{V}_{\mathbf{R}_1 + \mathbf{R}_0, \mathbf{R}_2 + \mathbf{R}_0; \mathbf{R}_3 + \mathbf{R}_0, \mathbf{R}_4 + \mathbf{R}_0}^{l_1, l_2; l_3, l_4}$ . What is normally used as an input parameter in model Hamiltonian studies or in the LDA+DMFT calculation or an RPA analysis of the instability is the left-hand side of Eq. (A3). It is clear that this interaction parameter depends on the choice of orbital (basis) and therefore its symmetry properties also depend on the basis. However, one can dress susceptibilities in the orbital basis; hence oscillator matrix elements only appear when closing the external legs of the dressed generalized susceptibilities to obtain observable susceptibilities.

### APPENDIX B: $P4/nmm$ SPACE GROUP

$P4/nmm$  is a nonsymmorphic group. It has eight point group operations (see Fig. 1 of main text). In addition to the eight point group operations, it also has an inversion,  $\{I|000\}$ . Together they form the 16 elements of  $P4/nmm$ . Note that this set is not closed under multiplication, and therefore, as is, it cannot form a group. However, if one defines the product modulo integer translations, then these 16 operations form a group.

The first eight symmetry operations can be generated by  $\{C_{2y}|0\frac{1}{2}0\}$  and  $\{C_{2a}|000\}$  as

$$\begin{aligned} \{C_{2y}|0\frac{1}{2}0\}\{C_{2a}|000\} &= \{C_{4z}^+|0\frac{1}{2}0\}, \\ \{C_{2a}|000\}\{C_{2y}|0\frac{1}{2}0\} &= \{C_{4z}^-|0\frac{1}{2}0\}, \\ \{C_{4z}^+|0\frac{1}{2}0\}\{C_{4z}^+|0\frac{1}{2}0\} &= \{C_{2z}|0\frac{1}{2}0\} + \{E|-1,0,0\}, \\ \{C_{2y}|0\frac{1}{2}0\}\{C_{2z}|0\frac{1}{2}0\} &= \{C_{2x}|0\frac{1}{2}0\} + \{E|-1,1,0\}, \\ \{C_{2a}|000\}\{C_{2z}|0\frac{1}{2}0\} &= \{C_{2b}|0\frac{1}{2}0\}, \end{aligned}$$

where the translational part  $\{E|-1,0,0\}$  will be dropped hereafter. Note that the  $b$  axis is in the Fe plane, so  $C_{2b}$  transfers the As ions from above to below and vice versa; hence a half-integer translation is required to bring the lattice into itself. The other eight operations can be obtained by applying inversion  $\{I|000\}$  on the previous operators.

### APPENDIX C: EFFECT OF NONSYMMORPHIC OPERATIONS

Let the basis set be  $\phi_{l_\alpha}(\mathbf{R}_j + \mathbf{r}_\alpha)$ . Here,  $\mathbf{R}_j$  denotes the position of the  $j$ th unit cell and  $\mathbf{r}_\alpha$  the position of the atom  $\alpha$  within the unit cell. The index  $\alpha$  can take four values, two of which are associated with Fe and two with As. Associated with each atom, there is a set of atomic orbitals denoted by  $l_\alpha$  where for a given  $\alpha$ ,  $l_\alpha$  can take several values. For example, if atom  $\alpha$  is an Fe atom, then  $l_\alpha$  can take five values corresponding to  $d$  orbitals while if it is an As atom, then  $l_\alpha$  can take three values corresponding to the three  $p$  orbitals. In the main text, the label  $l$  for the ‘‘orbital’’ labels both the atomic position and the orbital on the atom. Here we temporarily make the distinction between the atomic orbital and the position of the atom.

The action of  $\{g|\boldsymbol{\tau}\}$  is given by

$$\{g|\boldsymbol{\tau}\}\phi_{l_\alpha}(\mathbf{R}_j + \mathbf{r}_\alpha) = \phi_{l'_\alpha}(g\mathbf{R}_j + g\mathbf{r}_\alpha + \boldsymbol{\tau}), \quad (C1)$$

where  $(\alpha, l_\alpha)$  transfers to  $(\alpha', l'_\alpha)$  under the symmetry operation. The operations act not only on spatial coordinates, they also act on the atomic orbital itself. Take, for example,  $C_{2y}$  for  $g$ , then it would transform a  $p_z$  orbital of As into  $-p_z$  and would leave a Fe  $d_{xz}$  orbital unchanged. We also have to remember that the action of the operation  $g$  (that leaves the origin invariant) on the vectors  $\mathbf{R}_j$  and  $\mathbf{r}_\alpha$  is represented by the same  $3 \times 3$  matrix. We also have the result

$$g\mathbf{r}_\alpha + \boldsymbol{\tau} = \mathbf{r}_{\alpha'} + \mathbf{L}_\alpha, \quad (C2)$$

where  $\mathbf{r}_{\alpha'}$  is in the  $g\mathbf{R}_j$  unit cell and  $\mathbf{L}_\alpha$  is a real space lattice vector necessary to reach, from  $\mathbf{r}_{\alpha'}$ , the position  $g\mathbf{r}_\alpha + \boldsymbol{\tau}$  that may be in a unit cell adjacent to  $g\mathbf{R}_j$ . It depends on the original position  $\mathbf{r}_\alpha$ . The position  $\mathbf{r}_{\alpha'}$  will correspond to the same kind of atom, Fe or As, as the original one, but may be displaced in

the unit cell. A Fe atom originally on sublattice  $A$  may have moved to sublattice  $B$  for example; in other words,  $\alpha$  and  $\alpha'$  may be different, even if they both refer to an Fe atom.

The action of  $\{g|\boldsymbol{\tau}\}$  on the orbitals  $\phi_{l_\alpha}$  may be represented by a unitary matrix. Hence we may write

$$\{g|\boldsymbol{\tau}\}\phi_{l_\alpha}(\mathbf{R}_j + \mathbf{r}_\alpha) = U_g^{l_\alpha:l'_\alpha} \phi_{l'_\alpha}(g\mathbf{R}_j + \mathbf{r}_{\alpha'} + \mathbf{L}_\alpha), \quad (C3)$$

where here  $\alpha'$  is given and a summation over the different values  $l'_\alpha$  associated with atom  $\alpha'$  is implied.

We now move to Fourier transforms. We use the standard gauge because the Bloch Hamiltonian is periodic in this gauge. We comment on the other gauge whenever it is appropriate. We define

$$\phi_{\alpha l_\alpha}(\mathbf{k}) = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_j} e^{i\mathbf{k}\cdot\mathbf{R}_j} \phi_{l_\alpha}(\mathbf{R}_j + \mathbf{r}_\alpha). \quad (C4)$$

The action of  $\{g|\boldsymbol{\tau}\}$  on  $\phi_{\alpha l_\alpha}(\mathbf{k})$  follows if we also remember that under  $g$  (seen simply as a coordinate change, for example), the scalar product must be invariant so that  $g\mathbf{k}\cdot g\mathbf{R}_j = \mathbf{k}\cdot\mathbf{R}_j$ . We find

$$\begin{aligned} \{g|\boldsymbol{\tau}\}\phi_{\alpha l_\alpha}(\mathbf{k}) &= \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_j} e^{i\mathbf{k}\cdot\mathbf{R}_j} U_g^{l_\alpha:l'_\alpha} \phi_{l'_\alpha}(g\mathbf{R}_j + g\mathbf{r}_\alpha + \boldsymbol{\tau}) \\ &= \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_j} e^{i\mathbf{k}\cdot g\mathbf{R}_j} U_g^{l_\alpha:l'_\alpha} \phi_{l'_\alpha}(g\mathbf{R}_j + \mathbf{r}_{\alpha'} + \mathbf{L}_\alpha) \\ &= \frac{1}{\sqrt{N}} e^{-i\mathbf{k}\cdot\mathbf{L}_\alpha} \sum_{\mathbf{R}_j} e^{i\mathbf{k}\cdot(g\mathbf{R}_j + \mathbf{L}_\alpha)} U_g^{l_\alpha:l'_\alpha} \\ &\quad \times \phi_{l'_\alpha}(g\mathbf{R}_j + \mathbf{r}_{\alpha'} + \mathbf{L}_\alpha) \\ &= e^{-i\mathbf{k}\cdot\mathbf{L}_\alpha} U_g^{l_\alpha:l'_\alpha} \phi_{\alpha' l'_\alpha}(g\mathbf{k}). \end{aligned} \quad (C5)$$

Returning to the ‘‘orbital’’ notation of the main text, we combine  $\alpha$  and  $l_\alpha$  into a single large vector with index  $l \equiv (\alpha, l_\alpha)$ . The phase  $e^{-i\mathbf{k}\cdot\mathbf{L}_\alpha}$  can then be represented by a diagonal unitary matrix  $\mathbf{U}'_{g\mathbf{k},\mathbf{L}}$  that depends on  $g$  and also on the  $\alpha$  part of the index  $l$ . Similarly, using the new indices,  $U_g^{l_\alpha:l'_\alpha}$  can be rewritten so that all indices can be taken into account at once, leading, in vector notation, to

$$\{g|\boldsymbol{\tau}\}\phi(\mathbf{k}) = \mathbf{U}'_{g\mathbf{k},\mathbf{L}} \mathbf{U}_g \phi(g\mathbf{k}). \quad (C6)$$

### APPENDIX D: SYMMETRY CONSIDERATIONS FOR A MINIMAL TIGHT-BINDING HAMILTONIAN

In this section we further clarify the above symmetry concepts using a minimal Hamiltonian where we consider only Fe- $d_{x^2-y^2}$  and As- $p_z$  orbitals. The unit cell includes two Fe ions and two As ions; hence the Hamiltonian is a  $4 \times 4$  matrix. For the sake of simplicity, we consider a quasi-2D system while in reality there is a finite dispersion in the  $z$  direction. The unit cell vectors are  $\mathbf{a}_1 = (a, 0, 0)$  and  $\mathbf{a}_2 = (0, a, 0)$  and ion positions are  $\mathbf{r}_{A-\text{Fe}} = (-a/4, -a/4, 0)$ ,  $\mathbf{r}_{A-\text{As}} = (-a/4, a/4, -h)$ ,  $\mathbf{r}_{B-\text{Fe}} = (a/4, a/4, 0)$ , and  $\mathbf{r}_{B-\text{As}} = (a/4, -a/4, h)$ , where  $A/B$  label sublattices and the origin is on the inversion center.

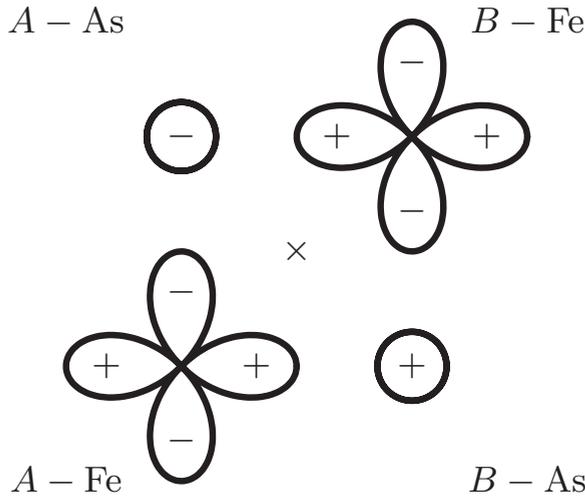


FIG. 4. Schematics of a FeAs trilayer unit cell with Fe- $d_{x^2-y^2}$  and As- $p_z$ . Note that for As- $p_z$  only the sign of the orbital's lobe near to the Fe-Fe plan is shown. The hopping matrix elements between Fe- $d_{x^2-y^2}$  and As- $p_z$  change sign by changing Fe ion. The cross shows the inversion center.

### 1. Minimal Hamiltonian and its symmetry properties

The Hamiltonian is given by  $\mathcal{H} = \sum_{\mathbf{k}} \phi_{\mathbf{k}}^{\dagger} \mathbf{H}_{\mathbf{k}} \phi_{\mathbf{k}}$ , where the basis set is

$$\phi_{\mathbf{k}} \equiv \{\phi_{A-\text{Fe}}(\mathbf{k}), \phi_{A-\text{As}}(\mathbf{k}), \phi_{B-\text{Fe}}(\mathbf{k}), \phi_{B-\text{As}}(\mathbf{k})\}^T. \quad (\text{D1})$$

With only nearest-neighbor hopping, the Hamiltonian, Eq. (9), in the standard gauge is given by

$$\begin{bmatrix} \epsilon_d & t(1+z_{y\mathbf{k}}^*) & 0 & t(1+z_{x\mathbf{k}}^*) \\ t(1+z_{y\mathbf{k}}) & \epsilon_p & -t(1+z_{x\mathbf{k}}^*) & 0 \\ 0 & -t(1+z_{x\mathbf{k}}) & \epsilon_d & -t(1+z_{y\mathbf{k}}) \\ t(1+z_{x\mathbf{k}}) & 0 & -t(1+z_{y\mathbf{k}}^*) & \epsilon_p \end{bmatrix}, \quad (\text{D2})$$

where  $t$  denote the nearest-neighbor hopping amplitude between As- $p_z$  orbitals and Fe- $d_{x^2-y^2}$ .  $\epsilon_d$ ,  $\epsilon_p$  and we defined

$$z_{x/y}(\mathbf{k}) \equiv \exp(-ik_{x/y}a). \quad (\text{D3})$$

From Fig. 4 one can see that the hopping amplitude between Fe and As has different signs for Fe-A and Fe-B. The Hamiltonian in the alternative gauge,  $\mathbf{H}'_{\mathbf{k}}$ , is obtained from Eq. (D2) by substituting  $t$  with  $t \exp[-i\mathbf{k} \cdot (\mathbf{r}_1 - \mathbf{r}_2)]$ , where  $\mathbf{r}_{1/2}$  denotes the orbital position.

The Hamiltonian  $\mathbf{H}_{\mathbf{k}}$  is trivially invariant under identity. Under inversion symmetry  $\{I|000\}$ , the wave vectors transform as  $(k_x, k_y, k_z) \rightarrow (-k_x, -k_y, -k_z)$ . The transformation matrix for the orbitals is given by

$$\mathbf{U}_I = \begin{bmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \\ 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \end{bmatrix}, \quad (\text{D4})$$

which gives  $\mathbf{U}_I^{\dagger} \mathbf{H}_{\mathbf{k}} \mathbf{U}_I = \mathbf{H}_{I\mathbf{k}} = \mathbf{H}(-k_x, -k_y, -k_z)$ . Note that the basis set transforms accordingly,  $\phi_{\mathbf{k}} \rightarrow \mathbf{U}_I \phi_{I\mathbf{k}}$ .

Another symmorphic generator of the group is  $\{C_{2a}|000\}$ . Under  $\{C_{2a}|000\}$  the wave vector transforms as  $(k_x, k_y, k_z) \rightarrow (k_y, k_x, -k_z)$  and the orbital space transformation matrix is

$$\mathbf{U}_{C_{2a}} = \begin{bmatrix} -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & -1 & 0 \\ 0 & -1 & 0 & 0 \end{bmatrix}. \quad (\text{D5})$$

Again the transformed Hamiltonian is  $\mathbf{U}_{C_{2a}}^{\dagger} \mathbf{H}_{\mathbf{k}} \mathbf{U}_{C_{2a}} = \mathbf{H}_{C_{2a}\mathbf{k}} = \mathbf{H}(k_y, k_x, -k_z)$ .

The last generator of the group is a nonsymmorphic symmetry operation, i.e.,  $\{C_{2y}|0\frac{1}{2}0\}$ . Under this symmetry operation  $(k_x, k_y, k_z) \rightarrow (-k_x, k_y, -k_z)$ . If one does not consider the extra phases induced by the half-integer translation, then in the truncated orbital space considered in this subsection, i.e., Fe- $d_{x^2-y^2}$  and As- $p_z$ , the orbital-space transformation matrix is the same as for inversion, Eq. (D4). Since the wave-vector transformation is different from that of inversion symmetry, one can immediately conclude that  $\mathbf{U}_{C_{2y}}^{\dagger} \mathbf{H}_{\mathbf{k}} \mathbf{U}_{C_{2y}}$  is not equal to  $\mathbf{H}_{C_{2y}\mathbf{k}} = \mathbf{H}(-k_x, k_y, -k_z)$ . Due to extra phases, in this case the correct orbital-space transformation matrix is

$$\mathbf{U}_{\{C_{2y}|0\frac{1}{2}0\}}(\mathbf{k}) = \begin{bmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -z_{y,\mathbf{k}} \\ z_{y,\mathbf{k}} & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \end{bmatrix}, \quad (\text{D6})$$

where the extra phases appear for As-A and Fe-B that leave the unit cell under this symmetry operation. Therefore,  $\mathbf{H}_{C_{2y}\mathbf{k}}$  is equal to  $\mathbf{U}_{\{C_{2y}|0\frac{1}{2}0\}}^{\dagger}(\mathbf{k}) \mathbf{H}_{\mathbf{k}} \mathbf{U}_{\{C_{2y}|0\frac{1}{2}0\}}(\mathbf{k})$  where, explicitly,

$$\begin{aligned} & \mathbf{U}_{\{C_{2y}|0\frac{1}{2}0\}}^{\dagger}(\mathbf{k}) \mathbf{H}_{\mathbf{k}} \mathbf{U}_{\{C_{2y}|0\frac{1}{2}0\}}(\mathbf{k}) \\ &= \begin{bmatrix} H_{33} & -z_{y,\mathbf{k}}^* H_{34} & z_{y,\mathbf{k}} H_{31} & -H_{32} \\ -z_{y,\mathbf{k}} H_{43} & H_{44} & -H_{41} & z_{y,\mathbf{k}} H_{42} \\ z_{y,\mathbf{k}} H_{13} & -H_{14} & H_{11} & -z_{y,\mathbf{k}} H_{12} \\ -H_{23} & z_{y,\mathbf{k}}^* H_{24} & -z_{y,\mathbf{k}}^* H_{21} & H_{22} \end{bmatrix}. \end{aligned} \quad (\text{D7})$$

Here,  $H_{ij}$ 's are matrix elements of  $\mathbf{H}_{\mathbf{k}}$  given in Eq. (D2). For example, consider the (1,2) component. It is given by  $-z_{y,\mathbf{k}}^* H_{34}$  where, using  $H_{34}$  from Eq. (D2), it is  $t z_{y,\mathbf{k}}^* (1 + z_{y\mathbf{k}}) = t(1 + z_{y\mathbf{k}}^*) = H_{12}(-k_x, k_y, -k_z)$ . The above equation is also valid in the alternative gauge if one replaces the matrix element of  $\mathbf{H}_{\mathbf{k}}$  with  $\mathbf{H}'_{\mathbf{k}}$  and employs the fact that the hopping matrix elements  $t \exp[-i\mathbf{k} \cdot (\mathbf{r}_1 - \mathbf{r}_2)]$  can be rewritten as  $t \exp[iC_{2y}\mathbf{k} \cdot C_{2y}(\mathbf{r}_1 - \mathbf{r}_2)]$ . We now have the transformation matrices for all of the generators of the group.

Finally, we are also interested in the fourfold symmetry that can be obtained as  $\{C_{4z}^+|0\frac{1}{2}0\}$ . Under this symmetry the wave vector changes as follows:  $(k_x, k_y, k_z) \rightarrow (-k_y, k_x, k_z)$  and the orbital space transforms as

$$\mathbf{U}_{\{C_{4z}^+|0\frac{1}{2}0\}} = \begin{bmatrix} 0 & 0 & -1 & 0 \\ 0 & 1 & 0 & 0 \\ -z_{x,\mathbf{k}} & 0 & 0 & 0 \\ 0 & 0 & 0 & z_{x,\mathbf{k}} \end{bmatrix}. \quad (\text{D8})$$

For  $\{C_{4z}^+|0\frac{1}{2}0\}$  the half-translation is in the  $y$  direction, so  $\mathbf{L} = \mathbf{a}_2$  for As and Fe ions on the  $B$  sublattice and the argument of the exponential is  $-ig\mathbf{k} \cdot \mathbf{L}$ , which gives  $z_{x,\mathbf{k}}$  for the extra phases.

## 2. Eigensystem of the minimal Hamiltonian

Now, let us consider the symmetry properties of the eigenstates. In this section we focus on fourfold symmetry in the  $k_z = 0$  plane. The eigenenergies of the Hamiltonian Eq. (9) are [17]

$$\begin{aligned} E_{\pm\pm,\mathbf{k}} &= \frac{(\epsilon_d + \epsilon_p)}{2} \pm \sqrt{\Delta^2 + (\lambda_{x\mathbf{k}} \pm \lambda_{y\mathbf{k}})^2} \\ &\equiv \frac{(\epsilon_d + \epsilon_p)}{2} \pm E'_{\pm\mathbf{k}}, \end{aligned} \quad (\text{D9})$$

where  $\Delta \equiv (\epsilon_d - \epsilon_p)/2$  and  $\lambda_{x/y\mathbf{k}}^2 \equiv t^2(1 + z_{x/y\mathbf{k}})(1 + z_{x/y\mathbf{k}}^*) = t^2[(1 + \cos k_{x/y}a)^2 + \sin^2 k_{x/y}a] = 4t^2 \cos^2(k_{x/y}a/2)$ . The eigenenergies have fourfold symmetry, i.e.,  $E_{\pm\pm,\mathbf{k}}$  ( $E'_{\pm\mathbf{k}}$ ) are invariant under  $(k_x, k_y) \rightarrow (k_y, -k_x)$  since under this transformation we have  $(\lambda_{x\mathbf{k}}^2, \lambda_{y\mathbf{k}}^2) \rightarrow (\lambda_{y\mathbf{k}}^2, \lambda_{x\mathbf{k}}^2)$ . As expected on general principles for nonsymmorphic space groups, the bands are degenerate on the BZ boundaries where  $k_x = \pm\pi/a$  or  $k_y = \pm\pi/a$  lead to vanishing  $\lambda_x$  or  $\lambda_y$ .

By diagonalizing Eq. (9), the expansion coefficients  $a_{n\mathbf{k}}^l = \langle \phi_{l\mathbf{k}} | \psi_{n\mathbf{k}} \rangle$  for  $E_{\mathbf{k}} = (\epsilon_d + \epsilon_p)/2 - (+)E'_{\pm\mathbf{k}}$  at a  $\mathbf{k}$ -point away from the diagonals are obtained as

$$\begin{aligned} |a_{-(+)\pm,\mathbf{k}}\rangle &\propto \left\{ -\frac{(\lambda_{x\mathbf{k}} \pm \lambda_{y\mathbf{k}})}{(\Delta + (-)E'_{\pm\mathbf{k}})} \exp(i\mathbf{k} \cdot \mathbf{r}_{A-Fe}), \pm \exp(i\mathbf{k} \cdot \mathbf{r}_{A-As}), \right. \\ &\quad \left. \frac{(\lambda_{y\mathbf{k}} \pm \lambda_{x\mathbf{k}})}{(\Delta + (-)E'_{\pm\mathbf{k}})} \exp(i\mathbf{k} \cdot \mathbf{r}_{B-Fe}), \exp(i\mathbf{k} \cdot \mathbf{r}_{B-As}) \right\}^T. \end{aligned} \quad (\text{D10})$$

For the  $\mathbf{k}$  points on the primary diagonal ( $k_x = k_y$ ) and secondary diagonal ( $k_x = -k_y$ ), we have the equalities  $\lambda_{x\mathbf{k}} = \lambda_{y\mathbf{k}}$  and  $E'_{-\mathbf{k}} = \Delta$ . The eigenstate with energy  $E_{+-,\mathbf{k}} = (\epsilon_d + \epsilon_p)/2 + E'_{-\mathbf{k}}$  for these  $\mathbf{k}$  points is

$$|a_{+-,\mathbf{k}}\rangle \propto \{\exp(i\mathbf{k} \cdot \mathbf{r}_{A-Fe}), 0, \exp(i\mathbf{k} \cdot \mathbf{r}_{B-Fe}), 0\}^T. \quad (\text{D11})$$

Finally, for the  $M = (\pm\pi/a, \pm\pi/a)$  the eigenvector of the  $E_{\mathbf{k}} = (\epsilon_d + \epsilon_p)/2 + E'_{+\mathbf{k}}$  may be chosen as

$$|a_{--,\mathbf{k}}\rangle \propto \left\{ i \frac{\sqrt{2}}{2} \frac{k_x}{|k_x|}, 0, i \frac{\sqrt{2}}{2} \frac{k_y}{|k_y|}, 0 \right\}^T. \quad (\text{D12})$$

The norms of the Bloch function, Eq. (D10), are equal to

$$\langle a_{-(+)\pm,\mathbf{k}} | a_{-(+)\pm,\mathbf{k}} \rangle = 2 + \frac{(\lambda_{x\mathbf{k}} \pm \lambda_{y\mathbf{k}})^2 + (\lambda_{y\mathbf{k}} \pm \lambda_{x\mathbf{k}})^2}{(\Delta - (+)E'_{\pm\mathbf{k}})^2}, \quad (\text{D13})$$

which has fourfold symmetry.

The Bloch states are gauge independent up to an overall phase. So, it does not matter whether we use the natural gauge or the alternative gauge. Even though eigenenergies

are invariant under fourfold symmetry, it is clear that the Bloch functions are not. However, the Bloch functions are auxiliary quantities and are not observable. On the other hand, an observable quantity such as the electron density  $n(\mathbf{r})$  is given by  $\sum_{n\mathbf{k}} f(\epsilon_{n\mathbf{k}}) |\psi_{n,\mathbf{k}}(\mathbf{r})|^2$  where  $f(\epsilon_{n\mathbf{k}})$  is the Fermi-Dirac distribution function. It has fourfold symmetry because the norm of the expansion coefficients, Eq. (D13), has this symmetry.

## APPENDIX E: SYMMETRY OF THE IN-PLANE RESPONSE FUNCTION OF THE MINIMAL HAMILTONIAN

Here we show explicitly that despite the fact that the generalized susceptibility Eq. (23) obtained from the minimal model Hamiltonian does not have  $C_{4z}$  symmetry for  $q_z = 0$ , the observable susceptibility at  $q_z = 0$  does. We need to find how the observable in-plane response function transforms under  $\{C_{4z}|0\frac{1}{2}0\}$  by inspecting the symmetry properties of Eq. (22), which includes the oscillator matrix elements. In this section, we assume that the applied field wave vector lies within the BZ and set  $\mathbf{G}, \mathbf{G}' = \mathbf{0}$ . In order to find how the in-plane  $\chi(\mathbf{q}, \nu)$  with  $q_z = 0$  transforms to  $\chi(C_{4z}\mathbf{q}, \nu)$  under  $\{C_{4z}|0\frac{1}{2}0\}$  symmetry, we need to transform each term in  $\chi(\mathbf{q}, \nu)$  to the corresponding term with  $(\mathbf{k}, \mathbf{q}) \rightarrow C_{4z}(\mathbf{k}, \mathbf{q})$ ,  $A - \text{Fe} \leftrightarrow B - \text{Fe}$ , and invariant As ions. Note that  $\epsilon_{n,\mathbf{k}} = \epsilon_{n,C_{4z}\mathbf{k}}$ , so we focus on the transformation properties of the expansion coefficients and the oscillator matrix elements in Eq. (22) and Eq. (23).

For example, consider the following contribution which includes intersite oscillator matrix elements between Fe sites and As:

$$\begin{aligned} a_{n,\mathbf{k}-\mathbf{q}}^{A-As*} a_{m,\mathbf{k}}^{A-Fe} a_{m,\mathbf{k}}^{B-As*} a_{n,\mathbf{k}-\mathbf{q}}^{B-Fe} \\ \times O_{\mathbf{k}}^{A-As, A-Fe}(\mathbf{q}) O_{\mathbf{k}}^{B-As, B-Fe}(-\mathbf{q}). \end{aligned} \quad (\text{E1})$$

We need to sum over all  $k_z$  to compute the response functions, but since  $k_z$  is invariant under  $C_{4z}$ , we present for simplicity the analysis for the  $k_z = 0$  case only. By comparing  $|a_{n,\mathbf{k}}\rangle$  and  $|a_{n,C_{4z}\mathbf{k}}\rangle$ , where  $C_{4z}(k_x, k_y) \rightarrow (-k_y, k_x)$ , one can verify that the expansion coefficients of a general in-plane  $\mathbf{k}$  point, Eq. (D10), satisfy

$$\begin{aligned} a_{n,\mathbf{k}}^{A-As} &= \exp(-ik_x a/2) a_{n,C_{4z}\mathbf{k}}^{A-As}, \\ a_{n,\mathbf{k}}^{A-Fe} &= -\exp(-ik_x a/2) a_{n,C_{4z}\mathbf{k}}^{B-Fe}, \\ a_{n,\mathbf{k}}^{B-As} &= \exp(ik_x a/2) a_{n,C_{4z}\mathbf{k}}^{B-As}, \\ a_{n,\mathbf{k}}^{B-Fe} &= -\exp(ik_x a/2) a_{n,C_{4z}\mathbf{k}}^{A-Fe}. \end{aligned} \quad (\text{E2})$$

The negative signs in the second and fourth equations in Eq. (E2) do not exist for momenta on the diagonals and the band corresponding to Eq. (D11). However, in our simple case, Eq. (E1) is identically zero for these momenta and band index because the expansion coefficients on As sites are zero; hence our discussion is valid in general. However, as we proceed to show momentarily, the existence of these negative signs is essential to recover the symmetry. Therefore, any perturbation that causes a nonzero correction to the As expansion coefficients in Eq. (E2) will break fourfold symmetry.

Under  $\{C_{4z}|0\frac{1}{2}0\}$  the oscillator matrix elements, Eq. (17), transform as [see Eq. (D8) and its discussion]

$$\begin{aligned} \{C_{4z}|0\frac{1}{2}0\} O_{\mathbf{k}}^{A-As, A-Fe}(\mathbf{q}) &\rightarrow -O_{C_{4z}\mathbf{k}}^{A-As, B-Fe}(C_{4z}\mathbf{q}), \\ \{C_{4z}|0\frac{1}{2}0\} O_{\mathbf{k}}^{B-As, B-Fe}(-\mathbf{q}) &\rightarrow -e^{iq_x a} O_{C_{4z}\mathbf{k}}^{B-As, A-Fe}(-C_{4z}\mathbf{q}), \end{aligned} \quad (\text{E3})$$

where we used passive rotation and the identity  $\phi_{\mathbf{k}}^{A/B-As}(C_{4z}^{-1}\mathbf{r}-\boldsymbol{\tau}) = \phi_{C_{4z}\mathbf{k}}^{A/B-As}(\mathbf{r})$  and  $\phi_{\mathbf{k}}^{A/B-Fe}(C_{4z}^{-1}\mathbf{r}-\boldsymbol{\tau}) = -\phi_{C_{4z}\mathbf{k}}^{B/A-Fe}(\mathbf{r})$ . The negative sign in the latter identity comes from the  $d_{x^2-y^2}$  orbital symmetry.

By replacing Eq. (E2) for the expansion coefficients and Eq. (E3) for the oscillator matrix element in Eq. (E1), one can

see that the signs and the extra phases cancel out and Eq. (E1) transforms as

$$\begin{aligned} a_{n, C_{4z}(\mathbf{k}-\mathbf{q})}^{A-As*} a_{m, C_{4z}\mathbf{k}}^{B-Fe} a_{m, C_{4z}\mathbf{k}}^{B-As*} a_{n, C_{4z}(\mathbf{k}-\mathbf{q})}^{A-Fe} \\ \times O_{C_{4z}\mathbf{k}}^{B-Fe, A-As}(C_{4z}\mathbf{q}) O_{C_{4z}\mathbf{k}}^{B-As, A-Fe}(-C_{4z}\mathbf{q}). \end{aligned} \quad (\text{E4})$$

Hence Eq. (E1) maps into the corresponding term in  $\chi(C_{4z}\mathbf{q}, \nu)$  under fourfold symmetry. Clearly, the symmetry properties of both the generalized susceptibility, Eq. (23) or Eq. (30), and the oscillator matrix elements should be considered to obtain an in-plane fourfold response function. On the other hand, the in-plane generalized susceptibility by itself is not invariant under four-fold symmetry.

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- [1] G. F. Bassani and G. P. Parravicini, *Electronic States and Optical Transitions in Solids* (Pergamon Press, New York, 1975).
- [2] H. Hosono and K. Kuroki, Iron-based superconductors: Current status of materials and pairing mechanism, *Phys. C (Amsterdam, Neth.)* **514**, 399 (2015).
- [3] J. T. Park, D. S. Inosov, A. Yaresko, S. Graser, D. L. Sun, Ph. Bourges, Y. Sidis, Yuan Li, J.-H. Kim, D. Haug, A. Ivanov, K. Hradil, A. Schneidewind, P. Link, E. Faulhaber, I. Glavatsky, C. T. Lin, B. Keimer, and V. Hinkov, Symmetry of spin excitation spectra in the tetragonal paramagnetic and superconducting phases of 122-ferropnictides, *Phys. Rev. B* **82**, 134503 (2010).
- [4] P. J. Hirschfeld, Using gap symmetry and structure to reveal the pairing mechanism in Fe-based superconductors, *C. R. Phys.* **17**, 197 (2016).
- [5] P. A. Lee and X.-G. Wen, Spin-triplet  $p$ -wave pairing in a three-orbital model for iron pnictide superconductors, *Phys. Rev. B* **78**, 144517 (2008).
- [6] V. Cvetkovic and O. Vafeek, Space group symmetry, spin-orbit coupling, and the low-energy effective Hamiltonian for iron-based superconductors, *Phys. Rev. B* **88**, 134510 (2013).
- [7] Y. Wang, T. Berlijn, P. J. Hirschfeld, D. J. Scalapino, and T. A. Maier, Glide-Plane Symmetry and Superconducting Gap Structure of Iron-Based Superconductors, *Phys. Rev. Lett.* **114**, 107002 (2015).
- [8] N. Hao and J. Hu, Topological Phases in the Single-Layer FeSe, *Phys. Rev. X* **4**, 031053 (2014).
- [9] J. Hu, Iron-Based Superconductors as Odd-Parity Superconductors, *Phys. Rev. X* **3**, 031004 (2013).
- [10] M. H. Christensen, J. Kang, B. M. Andersen, I. Eremin, and R. M. Fernandes, Spin reorientation driven by the interplay between spin-orbit coupling and Hund's rule coupling in iron pnictides, *Phys. Rev. B* **92**, 214509 (2015).
- [11] M. H. Fischer, Gap symmetry and stability analysis in the multiorbital Fe-based superconductors, *New J. Phys.* **15**, 073006 (2013).
- [12] N. E. Bickers, *Self-Consistent Many-Body Theory for Condensed Matter Systems* (Springer-Verlag, New York, 2004), Chap. 6, pp. 237–296.
- [13] R. Nourafkan, G. Kotliar, and A.-M. S. Tremblay, Correlation-Enhanced Odd-Parity Interorbital Singlet Pairing in the Iron-Pnictide Superconductor LiFeAs, *Phys. Rev. Lett.* **117**, 137001 (2016).
- [14] J. H. Tapp, Z. Tang, B. Lv, K. Sasmal, B. Lorenz, P. C. W. Chu, and A. M. Guloy, LiFeAs: An intrinsic FeAs-based superconductor with  $T_c = 18$  K, *Phys. Rev. B* **78**, 060505 (2008).
- [15] M. Aichhorn, L. Pourovskii, V. Vildosola, M. Ferrero, O. Parcollet, T. Miyake, A. Georges, and S. Biermann, Dynamical mean-field theory within an augmented plane-wave framework: Assessing electronic correlations in the iron pnictide LaFeAsO, *Phys. Rev. B* **80**, 085101 (2009).
- [16] R. Nourafkan, Nodal versus nodeless superconductivity in isoelectronic LiFeP and LiFeAs, *Phys. Rev. B* **93**, 241116 (2016).
- [17] It is straightforward to show that Eq. (9) satisfies the following identity:  $([\mathbf{H}_{\mathbf{k}}/t - (\epsilon_d + \epsilon_p)/2t\mathbf{1}]^2 - [(\epsilon_d - \epsilon_p)^2/4t^2 + \lambda_{x\mathbf{k}}^2 + \lambda_{y\mathbf{k}}^2]\mathbf{1})^2 = 4\lambda_{x\mathbf{k}}^2\lambda_{y\mathbf{k}}^2\mathbf{1}$ .