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S. Maiti [1], T.A. Maier[2], T. Böhm[3,4], R. Hackl[4] and P. J. Hirschfeld[1]

[1] *Department of Physics, University of Florida, Gainesville, FL 32611*
[2] *Center for Nanophase Materials Sciences and Computer Science and Mathematics Division,*
Oak Ridge National Laboratory, Oak Ridge, TN 37831-6494, USA
[3] *Fakultät für Physik E23, Technische Universität München, 85748 Garching, Germany and*
[4] *Walther Meissner Institute, Bavarian Academy of Sciences and Humanities, 85748 Garching, Germany*
(Dated: November 14, 2016)

In unconventional superconductors, understanding the form of the pairing interaction is the primary goal. In this regard, Raman spectroscopy is a very useful tool, as it identifies the ground state and also the subleading pairing channels by probing collective modes. Here we propose a general theory for multiband Raman response and identify new features in the spectrum that can provide a robust test for a pairing theory. We identify *multiple* Bardasis-Schrieffer type collective modes and connect the weights of these modes to the sub-leading gap structures within a microscopic pairing theory. The conclusions are completely general, and we apply our approach to interpret the B_{1g} Raman scattering in hole-doped $BaFe_2As_2$.

Introduction: Fe-based superconductors (FeSC) appear to display *s*-wave pairing, with an order parameter that may change sign between Fermi surface (FS) pockets[1–4]. Theoretical calculations based on spin fluctuations have found that the *d*-wave channel can be strongly competitive, and even argued that *d*-wave could become the ground state for sufficiently strong electron[5, 6] or hole doping[7]. The consequences of a competing pairing channel were explored by Bardasis and Schrieffer[8], who predicted the existence of a new collective mode corresponding to the phase fluctuations of the subdominant (d−wave) order parameter above the ground state (s−wave). An analogous simple calculation was performed by Devereaux and Scalapino[9] for a typical FeSC electronic structure with $s\pm$ symmetry of the ground state with anisotropic gaps. They showed that the mode frequency should depend on $1/u_d - 1/\tilde{u}_s$, where u_d is the *d*-wave coupling constant and $\tilde{u}_s$ is a renormalized *s*-wave coupling that depends on the angular form of the gap in the condensed state.

Such a mode (called a Bardasis-Schrieffer (BS) mode or particle-particle exciton), couples to the Raman probe, but was never observed in conventional superconductors. Recently, however, measurements on $Ba_{1-x}K_xFe_2As_2$[10, 11], $NaFe_{1-x}Co_xAs$[12], $Ba(Fe_xCo_{1-x})_2As_2$[13] found peaks in the B_{1g} polarization spectrum which were consistent with a collective mode. Although, in the latter two cases, these peaks were identified with an excitonic mode originating due to the proximity to the nematic phase[14], the observation of multiple peaks in the former system for dopings farther from the region dominated by nematic fluctuations is rather puzzling. We thus propose that the peaks observed in the K-doped system are more likely BS modes.

In this Letter, we provide a scheme to calculate the Raman-response within a microscopic pairing theory, using the same microscopic interactions that lead to pairing, and point out several details of the Raman-spectrum that were either not expected or not explained before. In particular, we show that in crystalline systems multiple BS modes exist and appear with a characteristic weight in the spectrum. We believe that such a theory is necessary to accurately calculate the observed Raman intensity.

In fact, every subleading pairing channel leads to a BS mode (there may be multiple resonances). In crystalline systems with unconventional electronic structure, there can be an interplay of several orthogonal form-factors in the subleading channel as well. This aspect, which can strongly influence the shape of the Raman spectrum, appears to have been neglected until now. Theoretically, the solution to the linearized gap equation for a variety of materials[2] indeed shows the relevance of more than one subleading eigenfunction within the *same* irreducible representation of the normal state symmetry group. In particular, a spin fluctuation pairing calculation for the $Ba_{1-x}K_xFe_2As_2$ system indicates that the system condenses in a (A_{1g}) $s_\pm$ state, and that at least two subleading B_{1g} harmonics have non-negligible eigenvalues. We will show that this situation allows for the existence of well resolved BS peaks in the spectrum whose spectral weight distribution, as obtained from theory, seems consistent with the experiment.

Here we present a general scheme to compute the Raman scattering intensity for a multiband model, including vertex corrections. We then apply the general formulation to two specific cases and illustrate all of the points discussed above. The advantage of this formalism lies in the fact that it is valid for any ground-state with any number of bands and it accounts for all collective modes through the vertex corrections, which removes the singularity of the Raman response at twice the gap edge.

Multiband Raman response in the B_{1g} channel: The intensity of the Raman response in a multiband sys-
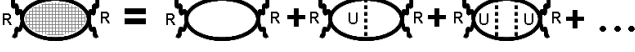
FIG. 1: The summation scheme for non-resonant Raman-scattering for the B_{1g} sector. The long-range Coulomb interaction does not affect this sector. Here U is shorthand for a generic residual interaction vertex in the pairing channel.
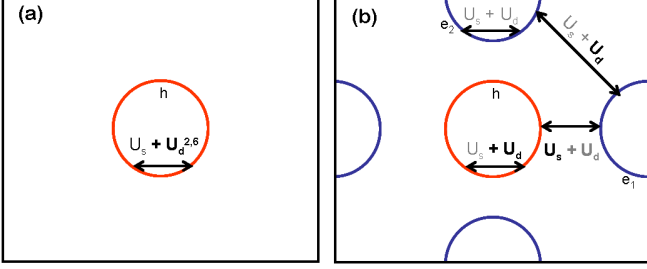

FIG. 2: Two toy models considered for illustration of the main results of this work: (a) One pocket around Γ-point with $s-$ and two $d-$interactions corresponding to 2θ and 6θ harmonics. (b) 3 pockets where only the interactions in the dark font are retained.

tem, in the non-resonant response limit, in the B_{1g} channel can be expressed as[15] $\chi_R(Q) \equiv \chi_R(\Omega, \vec{q}) \propto \sum_{a,b} \int dt e^{-i\Omega t} \langle \rho_a^R(t, \vec{q}) \rho_b^R(0, \vec{q}) \rangle \equiv \Pi^{RR}(Q)$, where ρ_a^R, in the non-resonant limit, is well approximated by the "Raman density" in the B_{1g} channel: $\sum_{\vec{k}} \gamma_{\vec{k}}^a c_{\vec{k}}^{a\dagger} c_{\vec{k}}^a$, where $\gamma_{\vec{k}}^a$ for band a in the B_{1g} channel is $\partial^2 \varepsilon_{\vec{k}}^a / \partial k_x^2 - \partial^2 \varepsilon_{\vec{k}}^a / \partial k_y^2$. Here Ω and $\vec{q}$ correspond to the shift in frequency and wavevector of the incident light. In metals, $q \ll k_F$, the Fermi wave vector, and will be set to zero in this work. We evaluate the above expression in the SC state using the summation scheme outlined in Fig. 1. To do so, we work with the Hamiltonian $H = H_0 + H_{int}$, where $H_0 = \sum_{\vec{k},\sigma,a} \varepsilon_{\vec{k}}^a c_{\vec{k},\sigma,a}^\dagger c_{\vec{k},\sigma,a} - \sum_{\vec{k}} \Delta_{\vec{k},a}^* c_{\vec{k},\uparrow,a} c_{-\vec{k},\downarrow,a} + c.c$ and

$$H_{int} = \sum_{a \neq b} U_{ab}^{(3)}(\vec{q}) c_{\vec{k},\alpha,a}^\dagger c_{\vec{k}'+\vec{q},\beta,a}^\dagger c_{\vec{k}',\beta,b} c_{\vec{k}+\vec{q},\alpha,b}$$
$$+ \sum_a U_{aa}^{(4)}(\vec{q}) c_{\vec{k},\alpha,a}^\dagger c_{\vec{k}'+\vec{q},\beta,a}^\dagger c_{\vec{k}',\beta,a} c_{\vec{k}+\vec{q},\alpha,a}.$$

This is the momentum dependent form of the interactions as modeled, e.g. in Ref. [16, 17] (the other interactions neglected here do not affect the main message of this work). We proceed by rotating the basis to the Nambu space with the spinor $\psi_{\vec{k}}^\dagger = (c_{\vec{k},\uparrow,1}^\dagger, c_{-\vec{k},\downarrow,1}, c_{\vec{k},\uparrow,2}^\dagger, c_{-\vec{k},\downarrow,2}, ...)$, where $1, 2...$ are the various bands. The interaction is recast as $H_{int} = \sum U^{\alpha\beta\gamma\delta}(\vec{q}) \psi_{\vec{k},\alpha}^\dagger \psi_{\vec{k}'+\vec{q},\beta}^\dagger \psi_{\vec{k}',\gamma} \psi_{\vec{k}+\vec{q},\delta}$. The explicit form of the interaction vertex is listed in the supplementary material(SM). Then, we need to evaluate

$$\Pi_{RR}(Q) = \sum_{a,b} \int_K \text{Tr} \left[\hat{R}_{a3} \hat{G}_K \hat{\Gamma}_3^{R_b} \hat{G}_{K+Q} \right], \quad (1)$$

$$\hat{\Gamma}_3^{R_a} = \hat{R}_{a3} - \sum_{c,m,n} U_{ac}^{nm} f_{\vec{k}}^n \int_{K'} f_{\vec{k}'}^{m*} \mathcal{M}_{ac} \cdot \hat{G}_{K'} \hat{\Gamma}_3^{R_c} \hat{G}_{K'+Q} \mathcal{M}_{ac}^\dagger,$$

where, $\hat{R}_{a3} = \sum_t f_{\vec{k}}^t c_a^t [\sigma_3 \otimes s_a]$; $\int_K \equiv T \sum_n \int \frac{d^2k}{(2\pi)^2}$; s_a is the band selector of the form diag $(0, ..., 1, 0, ...)$ (1 at the ath location); $\sigma_{1,2,3}$ are Pauli matrices in Nambu space; $\hat{G}$ is the bare Greens' function in Nambu space with elements $\hat{G}_a = [i\omega_n \sigma_0 - \epsilon_{\vec{k}}^a \sigma_3 - \Delta_{\vec{k},a} \sigma_1]^{-1}$ for band a; $\gamma_{\vec{k}}^a$ is expanded as $\sum_t f_{\vec{k}}^t c_a^t$, where $\{f_{\vec{k}}^t\}$ is a set of orthogonal functions within the B_{1g} sector; U_{ab}^{nm} is the projection matrix element of $U^{\alpha\beta\gamma\delta}$ for harmonics n, m for the interaction between bands a and b and $\mathcal{M}$ is a matrix that accounts for transformation to Nambu space (see SM). All matrices are $2N \times 2N$, where N is the number of bands and 2 is the Nambu space dimension. To proceed, it is necessary to introduce the other Nambu components: $\Gamma_i^{R_b}$ with $i = 1, 2$ in addition to $i = 3$, the solution for which is constructed as $\Gamma_i^{R_b} = \sum_{t,a} f_{\vec{k}}^t [\sigma_j \otimes s_a] \mathcal{K}_{ji}^{t,ab}$, where the matrix $\mathcal{K}_{ji}^{t,ab}$ is found after substituting for $\Gamma_i^{R_b}$ in Eq. 1. The response Π_{RR} is then given by

$$\Pi_{RR} = \sum_{a,d,t,t'} c^{t,a} \Pi_{3i}^{tt';a} \mathcal{K}_{i3}^{t',ad}, \text{ where} \quad (2)$$

$$\mathcal{K} = \left[1 + \frac{1}{4} [U_{pp}] \cdot [\tilde{\Pi} - \Pi] + \frac{1}{4} [U_{ph}] \cdot [\tilde{\Pi} + \Pi] \right]^{-1} [c],$$

$$\tilde{\Pi}_{ij}^{mt;b} = \int_{K'} f_{\vec{k}'}^{m*} f_{\vec{k}'}^t \text{Tr} \left[\mathcal{M}_{ab}^\dagger \cdot [\sigma_i]^a \mathcal{M}_{ab} \hat{G}_{K'} [\sigma_j]^b \hat{G}_{K'+Q} \right]. \quad (3)$$

Here $[U_{pp,ph}]$ is the coupling matrix in nambu⊗band⊗harmonic space and $\mathcal{K}$ and $[c]$ are matrices in nambu⊗band space, but a vector in harmonic space (see SM for examples); The subscript pp and ph for $[U]$ stand for its pairing and density channel projections. Further, $[\sigma_i]^a \equiv \sigma_i \otimes s_a$ and Π is the same as $\tilde{\Pi}$ but without the $\mathcal{M} - \mathcal{M}$ matrices.

The collective modes are contained in the poles of $\mathcal{K}$. While in general $\mathcal{K}$ is 4×4 in Nambu space, the singlet ground state which preserves time reversal symmetry decouples this into two 2×2 blocks: spin-amplitude and phase-density sectors. Since the BS modes are in the phase sector, we only deal with this 2×2 subspace in this work. The advantage of this formalism is that the Raman response is computed for a microscopic model where it is dressed by the same interactions that led to pairing. The microscopic problem provides the relevant number of harmonics H that effect the pairing problem and this approach to calculate Raman response then calls for computing the numbers Π^{nm} ($n, m \in \{1, ...H\}$) and carrying out a matrix inversion.

Connection to collective modes: It is well known that the collective modes in the sub-leading channel couple to the Raman response [9, 18]. However, several questions

remain: Are the poles in the Raman response always the same as the frequencies of collective modes? If there are multiple collective modes, how and with what spectral weight do they couple to the Raman probe?

Our formalism naturally provides answers to such questions. The poles of Raman response (Eq. 2) are contained in poles of $\mathcal{K} = [1 + [U_{pp}][\tilde{\Pi} - \Pi]/4 + [U_{ph}][\tilde{\Pi} + \Pi]/4]]^{-1}$. The collective mode in a general multiband superconductor, can be found, e.g. using the formalism in Ref. [19] and they are the poles of $[1 - [U][\Pi]/2]^{-1}$. Thus it is clear that, in general, the poles are *not* the same. However, if the interaction in the density channel is weak ($[U_{ph}] \to 0$), then one can show that in the pairing interaction sector, $\tilde{\Pi} \to -\Pi$ and we restore the collective mode result. This was also pointed out in Ref. [20] using a different scheme. In most works in the literature, the density channel has been neglected; for demonstration purposes, we shall do the same here. This precludes the appearance of a particle-hole exciton[20] in the Raman intensity analogous to the so-called "neutron reso-

nance" and does not affect any of our claims for the case of the B_{1g} Raman polarization. The answer regarding the weights of the various BS modes will be apparent in the following examples.

Simple toy models showing multiple BS modes: First we consider a Γ centered pocket, together with an s-wave BCS like ground state for the system with the order parameter Δ [23]. We then choose a competing SC B_{1g} channel with two harmonics via the interaction: $U_{\theta,\theta'} = 2U_{22}\cos 2\theta \cos 2\theta' + 2U_{66}\cos 6\theta \cos 6\theta' + 2U_{26}(\cos 2\theta \cos 6\theta' + \cos 6\theta \cos 2\theta')$. The gap structure has the form $\Delta_\theta = \sqrt{2}\Delta_2 \cos 2\theta + \sqrt{2}\Delta_6 \cos 6\theta$. The numbers we need to compute the B_{1g} Raman response are: $\Pi_{22,32}^{22,26,66}$, which in this model are: $\Pi_{22,32}^{22,66} = \Pi_{22,32}^{00} \equiv \Pi_{22,32}$ and $\Pi_{ij}^{26} = 0$. We also assume the harmonic decomposition of $\gamma_{\vec{k}}$ in terms of coefficients $c_{2,6}$. The Raman response, as computed in this formalism, is then given by

$$\Pi^{RR} = (c_2)^2 \left\{ \Pi_{33} - \frac{(\Pi_{23})^2 \left(U_{22}/2 + [U_{26}^2 - U_{22}U_{66}]\Pi_{22}/4 \right)}{\mathcal{D}} \right\} + (c_6)^2 \left\{ \Pi_{33} - \frac{(\Pi_{23})^2 \left(U_{66}/2 + [U_{26}^2 - U_{22}U_{66}]\Pi_{22}/4 \right)}{\mathcal{D}} \right\},$$
$$(4)$$

where $\mathcal{D} \equiv 1 - (U_{22} + U_{66})\Pi_{22}/2 - (\Pi_{22})^2(U_{26}^2 - U_{22}U_{66})/4$, zeroes of which correspond to peaks in the Raman spectrum. A simple exercise[21] shows us that this determinant is exactly the equation the determines the frequency of the BS modes. Important information about the pairing interaction can be more readily extracted if we rotate the interaction in the orthogonal basis functions provided by the eigen vectors of the pairing problem. When this rotation is done, $\{U_{22}, U_{26}, U_{66}\} \to \{\tilde{U}_1, 0, \tilde{U}_2\}$, $\{c_2, c_6\} \to \{\tilde{c}_1, \tilde{c}_2\}$; where $\tilde{c}_{1,2} = \int_\theta \left(\frac{\partial^2 \varepsilon}{\partial k_x^2} - \frac{\partial^2 \varepsilon}{\partial k_y^2} \right) |_{FS} \Delta_\theta^{(1,2)}$ are the overlap of the Raman vertex $\gamma_{\vec{k}}$ with the eigenvectors $\Delta_\theta^{(1,2)}$ (FS stands for projection on the Fermi surface). The response then takes the form $\Pi^{RR} =$

$$(\tilde{c}_1)^2 \left\{ \Pi_{33} - \frac{(\Pi_{23})^2}{\frac{2}{\tilde{U}_1} - \Pi_{22}} \right\} + (\tilde{c}_2)^2 \left\{ \Pi_{33} - \frac{(\Pi_{23})^2}{\frac{2}{\tilde{U}_2} - \Pi_{22}} \right\}.$$
$$(5)$$

Here $\Pi_{22} = \frac{2}{\tilde{U}_s} - 2\nu F(\Omega)$, where ν is the density of states at the FS and $F(\Omega) = \frac{(\Omega/2\Delta)\sin^{-1}(\Omega/2\Delta)}{\sqrt{1-(\Omega/2\Delta)^2}}$. The BS modes are solutions to $\nu F(\Omega) = -\frac{1}{\tilde{U}_{1,2}} + \frac{1}{\tilde{U}_s}$. These BS modes are weighted by $\tilde{c}_{1,2}$. Note that the weight of the BS mode goes to zero as it softens[22]. Fig. 3 displays the Raman response for various cases: when d-wave solution is not competing ($\tilde{U}_{1,2} > 0$, i.e. repulsive channel), there

are no collective modes. As more channels become competitive, collective modes begin to show up (Fig 3b-c). The weight of the collective modes are controlled by the electronic structure via the $\tilde{c}_1$ and $\tilde{c}_2$ coefficients. Since a microscopic theory for pairing is capable of providing the numbers $\tilde{U}_{1,2}$, $\tilde{c}_{1,2}$, augmenting such a theory with a calculation of Raman spectrum provides a much stronger testing ground for its validity. It is also clear from Fig. 3 that the harmonics of the interaction that 'host' the BS modes contribute very little to the 2Δ peak, an effect due to vertex correction and is analogous to what happens in A_{1g} sector [24]. There are contributions, however, to the 2Δ peak from other solutions that have an eigenvalue close to zero and the effect of vertex corrections is weak. This is shown explicitly in the next example.

We now consider an example where the electronic structure is more specific to FeSC. Such a model would consist of 1 hole and 2 electron pockets as shown in Fig. 2. To minimize the parameters and keep the calculations analytically tractable, we choose minimal interactions necessary to satisfy the symmetry requirement for the ground state of FeSC: we first restrict the interband interaction to be U_s in the s-channel, and further assume the following relations for the density of states at the Fermi level: $\nu_h = 2\nu_e \equiv \nu$. This results in an $s\pm$ state with $\Delta_h = -\Delta_e = \Delta$. We now choose the interaction in the B_{1g} channel with only U_d^{hh} and $U_d^{e_1 e_2}$
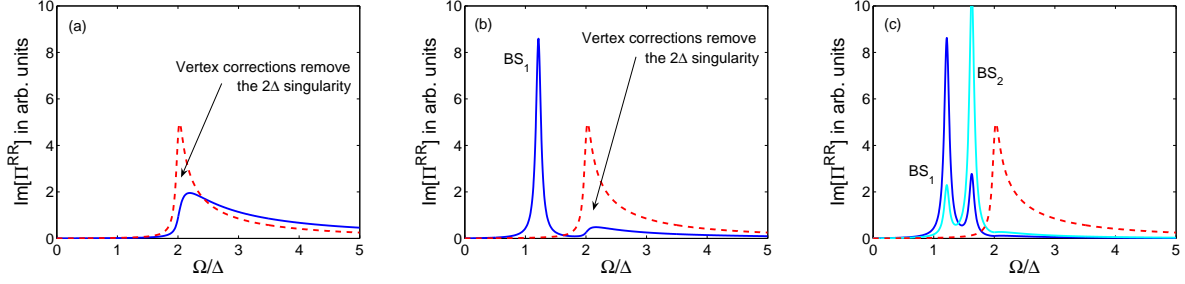
FIG. 3: Raman response for toy model with one pocket. The dashed red line in each case is the response in the absence of interactions. (a) Case where $d-$wave is not competitive: $\nu\tilde{U}_1 = 0.4$ and $\nu\tilde{U}_2 = 0.3$. (b) Case where only one $d-$wave solution is competitive: $\nu\tilde{U}_1 = -0.4$ and $\nu\tilde{U}_2 = 0.3$. (c) Case where both $d-$wave solutions are competitive: $\nu\tilde{U}_1 = -0.4$ and $\nu\tilde{U}_2 = -0.3$. The light and dark blue correspond to a band structure such that $\tilde{c}_1$ and $\tilde{c}_2$ are switched: this shows the connection of spectral weight of a Raman peak with the subleading eigenvectors. Here $\nu U_s = -0.5$, $\tilde{c}_1 = 0.6$ and $\tilde{c}_2 = 0.3$. A fermion lifetime of 0.05Δ was included to get the broadening.
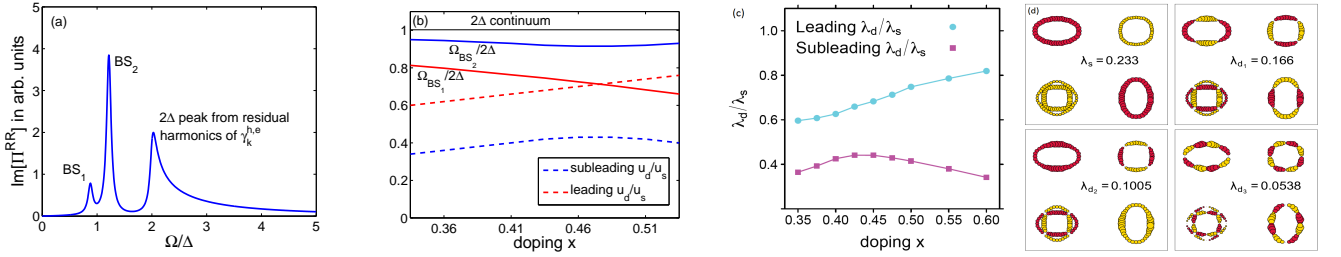

FIG. 4: (a)Raman response in the toy model with 3 pockets, with parameters $c^h = 0.2$, $c^e = 0.5$, $c^o = 0.3$, $\nu U_s = 0.5$, (broadening of 0.05Δ). The 2Δ feature in the spectrum remains because of non-zero c^o. (b) Correlating the evolution of the BS peaks by tuning the doping and the leading and subleading $d-$wave eigen-values ($U_d^{hh,e_1 e_2}$ are modeled with doping and chosen to mimic panel (c), which is 5-band calculation). (c) Calculated eigenvalues λ_{d_1} and λ_{d_2} in overdoped $Ba_{1-x}K_xFe_2As_2$ from RPA (see SM) with doping. (d) Corresponding eigenvectors plotted over the Fermi surface (the $\Gamma-$point has an inner and and outer pocket) for the ground-state $s_\pm$ (λ_{s_1}), and subsequent $d_{x^2-y^2}$-wave solutions($\lambda_{d_{1,2,3}}$) for $x = 0.55$. The symbol size is $\propto$ the gap size with red $= +$ and yellow $= -$.

components retained. This guarantees two competing subleading solutions: $[\Delta_h = \cos 2\theta; \; \Delta_{e_1,e_2} = 0]$ and $[\Delta_h = 0; \; \Delta_{e_1,e_2} = (1,-1)]$. We now define the overlaps of $\gamma^{h,e}(\vec{k})$ with harmonics in the interaction above to be c^h and c^e. The overlaps with remaining harmonics not dominant in the interaction are lumped under c^o. Following the same procedure as above and rotating the B_{1g} interaction in the pairing eigenvector basis, we find the Raman response to be analogous to Eq. 5 with $\tilde{c}_{1,2} \rightarrow c^{h,e}$ and an additional term, $(c^o)^2\{\Pi_{33}^h + \Pi_{33}^e\}$, from the residual harmonic content of $\gamma_{\vec{k}}^{h,e}$. This latter term is responsible for the 2Δ peak in the presence of BS modes, and represents the combined weight of all the Bardasis-Schrieffer modes of negligible strength, piled up around the two-particle continuum edge. In previous calculations, these contributions were neglected, so, if the collective modes were properly accounted for, the 2Δ peak was absent, in contrast to experiments[10–12]. This is the first explanation, to our knowledge, of this essential experimental feature. While the straightforward algebra is shown in SM, the Raman response is plotted

and explained in Fig.4.

Relevance to (Ba,K)Fe$_2$As$_2$: We now wish to apply this new understanding of the Raman spectrum to the overdoped region of BaKFe$_2$As$_2$. It is well known [7, 25, 26] in this system that higher hole doping makes the d-wave state competitive with the $s\pm$ ground state. We have carried out RPA calculations for the 5-orbital model for BaFe$_2$As$_2$ introduced in Graser et al.[27] using the usual spin- and charge-exchange interaction[28, 29]. As shown in Fig. 4c and d, these calculations find a leading $s\pm$ state and at least two subleading and competing B_{1g} states which are well resolved in energy[30]. Consequently, the insight from this work suggests appearance of two BS modes as a function of doping. Such a feature is reportedly seen in experiments[31] where the trend in the evolution of the peak positions correlates with the trend in eigenvalues just as shown for the toy model in Fig.4b.

Conclusions: In summary, we have provided a proof of principle method for using the details of the Raman spectrum, together with theory, to learn the details of the

pairing interaction in an unconventional superconductor. The calculation of the response with the full momentum structure of the interaction is outside the scope of this letter and will be considered in a more detailed future study. This formalism is readily generalizable to any ground-state symmetry and any number of bands. We have identified several features that help to better understand the Raman spectrum: a) there can be multiple Bardasis-Schrieffer modes in an s-wave superconductor; b) the overlap of the gap structure with the bare Raman vertex $\gamma_{\vec{k}}$ determines the weights of the modes c) incorporating the vertex corrections, we find that the 2Δ feature is suppressed and exists only due to the residual harmonics of the Raman vertex $\gamma_{\vec{k}}$ that are not involved in pairing.

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