Supplementary Figure 1. **Resistivity of** \( \text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2 \). Temperature dependence of resistivity in single crystals of \( \text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2 \).

Supplementary Figure 2. **Resonant Inelastic X-ray Scattering experimental geometry.** Experimental geometry of RIXS experiments.

**SUPPLEMENTARY NOTES 1**

In the intermediate coupling regime, the dynamical correlations are dominated by density-fluctuations. The effect of density-fluctuations to the electronic spectrum is included within a self-consistent self-energy calculation. The method used for the calculation is described below and is known as momentum-resolved density fluctuation (MRDF) theory\(^1\). The non-interacting Green’s function is obtained from the TB Hamiltonian \( \tilde{H}_k \) as

\[
\tilde{G}_0(k, \imath \omega_n) = \frac{1}{\imath \omega_n \tilde{I} - \tilde{H}_k},
\]

where \( \imath \omega_n \) is the Matsubara frequency for the fermions. The symbol \( \sim \) over a quantity
Supplementary Figure 3. **X-ray Absorption Spectroscopy (XAS) spectra at different doping levels.** Fe-L_{2,3} XAS of BaFe_{2}(As_{1-x}P_{x})_{2} for all the samples.

Supplementary Figure 4. **Full line fitting example.** Resonant Inelastic X-ray Scattering spectrum for BaFe_{2}As_{2} at (0.44, 0.0) depicted as black dots and fitting of the full emission line obtained employing the described formulas plotted as blue line.

reflected that it is a tensor. \( \tilde{I} \) is the identity matrix. The explicit form of \( G_{0} \) is

\[
\tilde{G}_{0,mn}(\mathbf{k}, \omega_{n}) = \sum_{\nu} \frac{\hat{\phi}_{k,m}^{\nu} \hat{\phi}_{k,n}^{\nu}}{\bar{\omega}_{n} - \xi_{\mathbf{k}}} ,
\]

Here \( \phi_{k,m}^{\nu} \) is the eigenstate for the \( \nu^{th} \) band \( (\xi_{n} u_{k}) \) projected onto the \( m^{th} \) orbital. The
Supplementary Figure 5. **Comparison of the Resonant Inelastic X-ray Scattering (RIXS) spectra in the whole energy loss range.** RIXS spectra for BaFe$_2$As$_2$, BaFe$_2$(As$_{0.72}$P$_{0.28}$)$_2$, and BaFe$_2$(As$_{0.48}$P$_{0.52}$)$_2$ at (0.44, 0).

Bare density fluctuation is obtained by the convolution of the Green’s function whose components can be written as

$$
\tilde{\chi}_{0,mn}^{st}(\mathbf{q}, \omega_p) = -\frac{1}{\Omega_{BZ}^\beta} \sum_{\mathbf{k},n} \tilde{G}_{0,mn}(\mathbf{k}, i\omega_n) \tilde{G}_{0,st}(\mathbf{k}+\mathbf{q}, i\omega_n + \omega_p),
$$

where $\beta = 1/k_B T$ with $k_B$ is the Boltzmann constant, and $T$ is the temperature. $\Omega_{BZ}$ is the phase space volume. $\mathbf{q}$, and $\omega_p$ are the momentum and frequency of the bosonic excitations created by density fluctuations. Without the self-energy correction, the base susceptibility can be evaluated easily by substituting the Green’s function from Eq. 2 in Eq. 3 and performing the Matsubara frequency summation over $\omega_n$ which yields,

$$
\tilde{\chi}_{0,mn}^{st}(\mathbf{q}, \omega_p) = -\frac{1}{\Omega_{BZ}} \sum_{\mathbf{k},\nu,\nu'} \phi_{\mathbf{k}+\mathbf{q},\nu} \phi_{\mathbf{k},\nu}^{\dagger} \phi_{\mathbf{k},\nu} \phi_{\mathbf{k},m}^{\dagger} \frac{f_{\mathbf{k}+\mathbf{q}} - f_{\mathbf{k}}}{\omega_p + i\delta - \xi_{\mathbf{k}+\mathbf{q}} + \xi_{\mathbf{k}}}
$$

Here $f_{\mathbf{k}}$ is the Fermi distribution function for the $\nu^{th}$ band. The interaction term includes multi-orbital components of the electronic interactions, including intra- $(U)$, inter-orbital $(V)$ Coulomb interactions, Hund’s coupling $(J)$ and pair-exchange $(J')$ terms:
Supplementary Figure 6. **Resonant Inelastic X-ray Scattering (RIXS) spectra at low energy loss for $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ along $(0, 0)\rightarrow(1, 1)$.** Momentum dependence of RIXS spectra along $(0, 0)\rightarrow(0.31, 0.31)$ for $x = 0.00, 0.15, 0.22, 0.28, 0.38, 0.48, \text{ and } 0.52$. The incoming photons are in $\pi$ polarization and the energy is tuned to the maximum of the Fe L$_3$ absorption edge. Experimental data are shown as black dotted lines, the background with the elastic peaks as purple dashed lines, and the magnetic peaks as green dashed lines. At low $q_{//}$ a fitting is unreliable, so no fitting was attempted.
Supplementary Figure 7. **X-ray Emission Spectroscopy (XES) experimental geometry.** Fe-K\(_\beta\) XES experimental geometry.

Supplementary Figure 8. **Fe-K\(_\beta\) XES and difference spectra for BaFe\(_2\)(As\(_{1-x}\)P\(_x\))\(_2\).** K\(_\beta\) XES for \(x = 0.00, 0.15, 0.22, 0.28, 0.38,\) and 0.52 (a-f as black lines) and reference spectrum of FeCrAs (a-f as red lines). The IAD is displayed as grey line. g Summary of IADs for all the samples.
Supplementary Figure 9. Fermi surface of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ for $x = 0.00$, 0.28, and 0.52. Calculated Fermi surfaces for BaFe$_2$(As$_{1-x}$P$_x$)$_2$ for $x = 0.00$, 0.28, and 0.52. As a black arrow, we indicate the nesting wave-vector.

\[
H_{int} = \sum_{\mathbf{k}_1-\mathbf{k}_4} \left[ \sum_{m,\sigma} U_{m} c_{\mathbf{k}_1\sigma}^m c_{\mathbf{k}_2\sigma}^m c_{\mathbf{k}_3\sigma}^m c_{\mathbf{k}_4\sigma}^m + \right.
\sum_{m<n,\sigma} \big( V_{mn} c_{\mathbf{k}_1\sigma}^m c_{\mathbf{k}_2\sigma}^m c_{\mathbf{k}_3\sigma}^n c_{\mathbf{k}_4\sigma}^n + (V - J_{mn}) c_{\mathbf{k}_1\sigma}^m c_{\mathbf{k}_2\sigma}^m c_{\mathbf{k}_3\sigma}^n c_{\mathbf{k}_4\sigma}^n \big) + \left. \sum_{m<n,\sigma} \big( J_{mn} c_{\mathbf{k}_1\sigma}^m c_{\mathbf{k}_2\sigma}^n c_{\mathbf{k}_3\sigma}^m c_{\mathbf{k}_4\sigma}^n + J'_{mn} c_{\mathbf{k}_1\sigma}^m c_{\mathbf{k}_2\sigma}^n c_{\mathbf{k}_3\sigma}^m c_{\mathbf{k}_4\sigma}^n + h.c. \big) \right]\]  

(5)

Here $c_{\mathbf{k}_1\sigma}^m$ ($c_{\mathbf{k}_1\sigma}^m$) is the creation (annihilation) operator for an orbital $m$ at momentum $\mathbf{k}_1$ with spin $\sigma = -\sigma = \uparrow$ or $\downarrow$. In the multiorbital spinor, the above interacting Hamiltonian can be expressed in the interaction tensor $\tilde{U}_{s/c}$, where the subscripts ‘$s$', ‘c' stand for spin and charge susceptibilities. The non-zero components of the $\tilde{U}_{s/c}$ tensor are given by

\[
\tilde{U}_{s,mn} = U, \quad \tilde{U}_{s,mm} = J, \quad \tilde{U}_{s,mm} = \frac{1}{2}J + V, \quad \tilde{U}_{c,mm} = J', \quad \tilde{U}_{c,mm} = \frac{3}{2}J - V, \quad \tilde{U}_{c,mm} = J'.
\]  

(6)

We have removed the orbital indices in all the interaction terms ($U$, $V$, $J$, $J'$). Within the random phase approximation, spin and charge channels become decoupled. The collec-
tive many-body corrections to the density-fluctuation spectrum can be written in matrix representation: 
\[ \tilde{\chi}_{c/s}(q, \omega_p) = \tilde{\chi}_0(q, \omega_p)[\tilde{T} \pm \tilde{U}_{c/s}\tilde{\chi}_0(q, \omega_p)]^{-1}, \] for the charge and spin parts, respectively. \( \tilde{\chi}_0(q, \omega_p) \) is the bare susceptibility tensor, whose components are given in Eq. 4 above, defined in the same basis as the interactions are defined. The interaction parameters are not parameterized individually. Rather, we estimate the components of the interaction tensor \( \tilde{U}_s \) from the Kanamori criterion as 
\[ \tilde{U}_{st}^{mn} \leq (\max[\tilde{\chi}_{st}^{mn}(q, \omega_p)])^{-1}. \] We find strong orbital dependence of the interaction parameters, such as \( U = 2, 2.2, 3.2, 2.5, 4.3 \) for all d-orbitals listed accordingly in the TB model, while \( V \approx U, J \approx U/2, \) and \( J' \approx J/2. \) \( \tilde{U}_c = \tilde{U}_s. \) We find little doping dependence of \( U \) and thus ignored it.

Finally, the density-fluctuation potential, applicable to the electronic states is calculated via the fluctuation-exchange approximation as
\[ \tilde{V}_{i,mm}^{st}(q, \omega_p) = \eta_i \left[ \tilde{U}_i \tilde{\chi}_i \tilde{U}_i \right]_{mn'}^{st}. \] (7)

Where \( i \) stands for spin (\( i = 1 \)) and charge (\( i = 2 \)) components, and \( \eta_1 = 3, \) and \( \eta_2 = 1. \) The corresponding self-energy is then calculated as
\[ \Sigma_{i,mm}(q, \omega) = \frac{1}{\Omega_{BZ}} \sum_{q,\nu,\nu'} \int_0^\infty \frac{d\omega_p}{2\pi} \tilde{V}_{i,mm}^{st}(q, \omega_p) \Gamma_{\nu,mm}^{st}(q, q) \left[ \frac{1 - f_{\nu, q} + n_p}{\omega_p + i\delta - \xi_{k-q}^{\nu} - \omega} + \frac{f_{\nu, q} + n_p}{\omega_p + i\delta - \xi_{k-q}^{\nu} + \omega} \right]. \] (8)

where \( n_p \) is the bosonic distribution function for frequency \( \omega_p. \) The vertex correction \( \Gamma_{\nu,mm}^{st}(q, q) \) encodes both the eigenstates of the \( \nu \)th band and the real part of the self-energy as
\[ \Gamma_{\nu,mm}^{st}(q, q) = \phi_{k-q,s}^{\nu} \phi_{k-q,s}^{\nu\dagger} \left( 1 - \frac{\partial \Sigma_{mm}^{\nu}(q, \omega)}{\partial \omega} \right)_{\omega=0}^{-1}. \] (9)

With the self-energy correction, the dressed Green’s function tensor becomes \( \tilde{G}^{-1}(q, \omega) = \tilde{G}_0^{-1}(q, \omega) - \tilde{\Sigma}(q, \omega). \) With the self-energy correction, the bare susceptibility cannot be calculated as simply as done in Eq. 4. Here we use a spectral representation as
\[ \tilde{G}(q, i\omega_n) = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{\tilde{A}(q, \omega')}{i\omega_n - \omega'}. \] (10)
where \( \tilde{A}(k, \omega') \) is the spectral function defined as \( \tilde{A}(k, i\omega_n) = -\text{Im}\tilde{G}(k, i\omega_n)/\pi \). By substituting Eq. 10 into Eq. 3, and doing a Matsubara summation, we get

\[
\tilde{\chi}_{st}^{0, mn}(q, \omega_p) = -\frac{1}{\Omega_{BZ}} \sum_k \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega''}{2\pi} \tilde{A}_{mn}(k, \omega') \tilde{A}_{st}(k+q, \omega'') f(\omega') - f(\omega'') \omega_p + i\delta - \omega' + \omega''
\]

(11)

Similarly, Eq. 8 is also modified accordingly. Eq. 11 gives the self-energy corrected bare density fluctuation spectrum. This self-energy corrected susceptibility gives a better description of the RIXS data as explained in the main text, compared to a simple RPA susceptibility without the self-energy correction.

**SUPPLEMENTARY NOTES 2**

In Supplementary Figure 10 we report the dynamical spin susceptibility calculated in the DFT-RPA (a) and MRDF (b) framework (we indicate with \( \Sigma \) the self energy). The equivalent dynamical charge susceptibility is drawn in Supplementary Figure 11. The color scale is kept the same for all the color plots in order to show the contrast in intensity. As written in the main text the local interactions, which are enhanced in MRDF, decrease the energy of the spin excitations and increase their intensity (see Supplementary Figure 10a and b). The intensity ratio of \( \chi_s \) and \( \chi_c \) shows that the former dominates the latter in agreement with a previous work performed on electron doped NaFeAs\(^2\). This is extremely evident especially in the color plot involving the MRDF theory as reported in Supplementary Figure 10 and 11.

It is informative to compare the RIXS signal with \( \chi_s \) on a qualitative level since the matrix elements have been neglected. In Supplementary Figure 10, we overlay with black dots including error bars the maximum of the spin excitations peak of our RIXS measurements with the theoretical calculations of \( \chi_s \). As seen in Supplementary Figure 10, there is no agreement between the DFT-RPA framework and the experimental data in terms of magnetic bandwidth. Additionally the DFT-RPA calculations do not capture the development observed with doping which predicts a softening rather than an hardening as observed in Supplementary Figure 10a. The MRDF calculations instead capture the magnetic bandwidth of the experimental data and the changes observed with doping as displayed in Supplementary
Supplementary Figure 10. **Comparison between Resonant Inelastic X-ray Scattering (RIXS) experiments and spin susceptibility calculations for BaFe$_2$(As$_{1-x}$P$_x$)$_2$.** a Color plot: Spin susceptibility ($\chi_s$) calculated by means of DFT-RPA. Black dots with error bars: Maximum of the spin excitations’ peak detected by RIXS experiments. b Color plot: Spin susceptibility ($\chi_s$) calculated by means of self-energy ($\Sigma$) corrected MRDF calculations. Black dots with error bars: Maximum of the spin excitations peak detected by RIXS experiments.

Figure 10.

For completeness, we also report a comparison between the RIXS experimental data and the calculations of $\chi_c$ in Supplementary Figure 11a and b for the DFT-RPA and MRDF, respectively. The calculations performed in the DFT-RPA do not agree with the data, whereas a better agreement can be observed in the MRDF shown in Supplementary Figure 11b. However the intensity of $\chi_s$ is an order of magnitude higher than $\chi_c$ indicating that most of the spectral weight observed in our RIXS experiments has to be ascribed to spin rather than charge excitations.
Supplementary Figure 11. **Comparison between Resonant Inelastic X-ray Scattering (RIXS) experiments and charge susceptibility calculations for BaFe$_2$(As$_{1-x}$P$_x$)$_2$.**

*Color plot: Charge susceptibility ($\chi_c$) calculated by means of Density Functional Theory (DFT)-Random Phase Approximation (RPA). White dots with error bars: Maximum of the spin excitations' peak detected by RIXS experiments.**

*Color plot: Charge susceptibility ($\chi_c$) calculated by means of self-energy ($\Sigma$) corrected Momentum Resolved Density Fluctuations theory (MRDF). White dots with error bars: Maximum of the spin excitations peak detected by RIXS experiments.*
**SUPPLEMENTARY REFERENCES**
