SLAC-PUB-16199

Supplementary Information for "Significant T_c enhancement in FeSe films on $SrTiO_3$ due to interfacial mode coupling"

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1 Growth and measurement methods

The single unit cell and multiple unit cell FeSe films in this study were grown on high quality single crystal 0.05%-wt Nb doped $SrTiO_3$ substrates (Shinkosha STEP substrates). The substrate was cut into 5 mm x 5 mm squares with a diamond saw and subsequently cleaned ultrasonically. No other surface preparation was necessary for these substrates. The cut substrates were mounted with silver paste onto a molybdnem sample holder. The silver paste was used to

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This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-76SF00515.

promote uniform heating of the substrate. The substrates were then introduced into our MBE chamber maintained at a base pressure of 3x10^{-11} Torr. The substrates were degassed at 450 °C for one hour and then slowly ramped up in temperature (approx 25 °C per minute) until a surface reconstruction was observed. The substrates were then annealed at 20 °C greater than the surface reconstruction temperature for 15 minutes and then lowered to the growth temperature at the same rate. The surface reconstruction was typically observed at 830 °C as measured with a pyrometer (assuming an emissivity of $\epsilon = 0.7$). The silver paste had minimal outgassing and growths typically occured at pressures better than 1x10^{-10} Torr. Ultrahigh purity selenium (99.999%) was evaporated from an effusion cell with a thermal cracking insert (Createc) while iron (99.995%) was evaporated from a 2 mm rod using an electron beam evaporator (Specs). The selenium evaporation rate of 0.13 Å/s was measured with a water cooled quartz crystal monitor, with an effusion cell temperature of 154.5 °C and a cracking insert temperature of 280 °C. The iron evaporation rate was maintained at 0.04 Å/s by utilizing a flux controller located at the end of the pocket. The substrate temperature was held at 380 °C during growth as measured with the pyrometer. These growth conditions resulted in high quality thin films with single unit cell coverage achieved within 30 seconds as observed by RHEED oscillations as shown in Extended Data (ED) Fig. 1. The films were subsequently annealed at 450 °C for four hours immediately after growth. The films were transported to the Stanford Synchrotron Radiation Lighsource ARPES beamline 5-4 via a vacuum suitcase with base pressure of 5x10^{-10} Torr. Several of the films were stored up to 4-6 days prior to measurement at the beamline. Clear photoemission spectra were typically observed after removal from the suitcase, but were often improved by a second four hour anneal at 450 °C while at the beamline.

All ARPES data taken at the beamline were with an energy resolution of 8 meV and angular resolution of $0.3°$. Unless otherwise noted, spectra taken at the synchrotron used a photon energy of 24 eV.

2 Additional data on films

The raw ARPES spectra are plotted in ED Fig. 2 along with their second derivatives. Films measured were 1 unit cell (1UC), 1.7UC, 2UC and 30UC in thickness. A high-temperature scan of the 30UC film is plotted in ED Fig. 2e, 2j, 2o, and 2t. The labelling of the replica bands at M used in the main text is presented again in ED Fig. 2p.

The 2UC and 30UC films have qualitatively similar band structures, but are dramatically different from the 1UC film. In the multi-UC films, the hole bands at Γ (ED Fig. 2c-d) are much closer to E_F, with the band tops crossing E_F, compared to the case of 1UC, where the band top is 80 meV below E_F. At the M point (ED Fig. 2m-n), the multi-UC band structure is more complicated, with two apparent hole-like bands at low temperature. This is in contrast with the 1UC film M point, where a single hole-like band exists, and two nearly degenerate electron bands cross E_F (see section 8). These differences are consistent with a previous report.[4] This behavior in the multi-UC film is reminiscent of the band shift observed in the iron pnictides,[28] which is associated with the tetragonal to orthorhombic structural transition.

With increasing temperature, we observe that the band splitting of the 30UC film begins to disappear. At 140 K (ED Fig. 2o, t), the band structure recovers to that of the normal state, with a single hole-like band at M, electron bands crossing E_F at M, and hole-like bands at Γ. Such evolution of the electronic structure suggests the existence of a tetragonal to orthorhombic structural transition in the multi-UC film, consistent with the observation of such a structural transition in bulk FeSe.[29] We also note that we do not observe evidence of band folding, suggesting the lack of long range magnetic order in multi-UC films, consistent with the lack of such an order in bulk FeSe. Finally, we note that from the high temperature normal state band structure of 30UC (ED Fig. 2e, j, o, t), the electron band bottom is slightly below E_F at M and the hole band top is slightly above E_F at Γ, suggesting that the doping level is close to zero, in

contrast to the case of 1UC, where heavy electron doping is observed.

We include a 1.7UC film to demonstrate the dramatic differences in spectra from the 2UC and the 1UC film. We find the 1.7UC spectra to be a superposition of the single UC and multi-UC films. The crossing of bands without hybridization indicates the film is composed of spatially separated regions of 1UC and 2UC films. In particular, for the 1.7UC film the 1UC bands still show replicas, while the 2UC bands do not (see ED Fig. 2q).

To rule out the possibility of quantum well states as the cause of the replica bands, we consider the following four facts: (1) The replica band ($\mathbf{A}'$) back-bends around the same momentum as the main band ($\mathbf{A}$); (2) The replica band ($\mathbf{A}'$) terminates at a similar momentum as the main band ($\mathbf{A}$) at an energy well below E_F; (3) The replica bands have not been seen in LDA calculations for FeSe on STO, suggesting that they are not band structure effects;[30,31] (4) Instead of following a well-understood evolution as a function of layer thickness, the replica bands disappear for FeSe films thicker than 1UC. As all 4 of these facts contradict the expected behavior for quantum well states,[17,18] we rule out such a scenario.

The replica bands $\mathbf{C}$ and $\mathbf{D}'$ presented in the main text are only partially resolved, as they intersect other bands with much larger spectral weight. However, for similar reasons stated for $\mathbf{A}'$ and $\mathbf{B}'$, both bands are also attributed to a phonon shakeoff mechanism, with $\mathbf{C}$ likely due to a different phonon branch. Because of the extremely low spectral weight of these bands we instead focus on $\mathbf{A}'$ and $\mathbf{B}'$ for our quantitative analyses below.

The temperature dependence of the 1UC film second derivative spectra is plotted in ED Fig. 3. The replica bands demonstrate a clear back-bending at low temperatures just like the main bands. Furthermore, we clearly observe the replica bands up to 90 K, as seen in ED Fig. 3e, with hints of the replica bands observed at 120 K, shown in ED Fig. 3f. Such temperatures are significantly higher than the gap-opening temperature, where Cooper pairs form. Higher temperatures resulted in spectra that were too broad to observe a replica band, which is dominated

by a large background, discussed in the next section.

3 Fitting the ARPES intensity spectra

In order to obtain an estimate of the electron-phonon (e-ph) coupling in the system, one needs to extract the intensity ratio between the main band and replica band. As plotted in ED Fig. 4a, we see at the M point that the band peaks sit on top of a large, non-monotonic background. Several models give reasonable fits to the data and we show two extremes: a spline interpolation background and a Shirley background,[32] both plotted in ED Fig. 4a. The blue circles indicate the points used to determine the spline. ED Fig. 4b plots the fit to the data after the spline fit is subtracted, with the individual peaks plotted separately for clarity. The fitted peak intensity ratio between $\mathbf{A}'$ and $\mathbf{A}$ is 1 to 6.2. Similarly, ED Fig. 4c plots the fit after subtraction of the Shirley background. The fitted binding energy range is from -0.32 eV to 0.03 eV. Individual peaks are also plotted. In this scenario the intensity ratio between $\mathbf{A}'$ and $\mathbf{A}$ is much larger and closer to 1 to 1. As discussed below, a larger intensity ratio implies a larger e-ph coupling interaction strength. For the sake of argument in providing a lower bound on the e-ph coupling strength, we choose the spline fit, which provides the smallest replica band to main band ratio.

4 A simple model for the electron-phonon interaction

Our experimental data indicates that the coupling between the FeSe electrons and the STO phonon is sharply peaked at zero momentum transfer. A natural question is what causes such strong localization in momentum space.

In the following we consider a single model that can give rise to the peculiar e-ph coupling described above. Let $\delta h(x, y)$ be the polar displacement (in the direction perpendicular to the interface ($\hat{z}$)) of the STO ions near the interface, and x, y are the coordinates parallel to the interface. Such displacement creates excess dipole moments situated at a distance h_0 away

from the FeSe electrons,

$$\delta p_z(x,y) = q_{\text{eff}} \delta h(x,y). \tag{1}$$

In the presence of anisotropic dielectric constants $\epsilon_\parallel$ (parallel to the interface) and $\epsilon_\perp$ (perpendicular to the interface) such dipole moments exert an electrostatic potential

$$\delta\Phi(x,y) = \frac{\epsilon_\parallel q_{\text{eff}} h_0}{\epsilon_\perp^{3/2}} \int dx'dy' \frac{\delta h(x',y')}{\left(\frac{\epsilon_\parallel}{\epsilon_\perp} h_0^2 + (x-x')^2 + (y-y')^2\right)^{3/2}} \tag{2}$$

Fourier transforming the above equation we obtain

$$\delta\Phi(\mathbf{q}_\parallel) = \sqrt{\frac{\epsilon_\parallel}{\epsilon_\perp}} \left(\frac{2\pi q_{\text{eff}}}{\sqrt{\epsilon_\perp}}\right) e^{-\left(|\mathbf{q}_\parallel| h_0 \sqrt{\epsilon_\parallel/\epsilon_\perp}\right)} \delta h(\mathbf{q}_\parallel) \tag{3}$$

where $\mathbf{q}_\parallel$ is the wavevector parallel to the interface. As a result the e-ph interaction is given by

$$\sum_q \rho(\mathbf{q}) \delta\Phi(\mathbf{q}), \tag{4}$$

where

$$\delta\rho(\mathbf{q}) = \sum_{\mathbf{k},\sigma} c^\dagger_{\mathbf{k}+\mathbf{q},\sigma} c_{\mathbf{k},\sigma} \tag{5}$$

is the electron density operator. The dielectric constant contains the contribution from both STO and the FeSe film. The motion of the FeSe electron is confined to directions parallel to the interface and hence can screen the electric field parallel but not perpendicular to the interface. The latter consideration, plus the fact that the screening from cubic STO is expected to be fairly isotropic, leads us to expect $\epsilon_\parallel$ to be significantly bigger than $\epsilon_\perp$. Two important enhancement effects are resulted: (1) the strength of the electron-phonon coupling is enhanced by $\sqrt{\epsilon_\parallel/\epsilon_\perp}$; (2) h_0 is enhanced by a factor $\sqrt{\epsilon_\parallel/\epsilon_\perp}$ resulting in the e-ph coupling function more sharply peaked at $\mathbf{q} = 0$ (momentum range $q_0 = \sqrt{\epsilon_\perp/\epsilon_\parallel}/h_0$). Such enhancement of apparent length scale in highly anisotropic dielectric environment has been discussed in polyacetylene.[33] In the following we shall use this form of coupling function.

5 A simple model for the replica bands

To understand the physical origin of the replica band we start with a simple toy model. Since the $\mathbf{q} = 0$ phonon couples most strongly with the electron we shall retain only this phonon mode. The model Hamiltonian reads

$$H = \sum_{\mathbf{k},\sigma} \epsilon(\mathbf{k}) n_{\mathbf{k},\sigma} + g(b^\dagger + b)\delta N + \hbar\Omega_0 \left(b^\dagger b \ + \frac{1}{2} \right) \tag{6}$$

where $\delta N = \sum_{\mathbf{k},\sigma} n_{\mathbf{k},\sigma}$ and $n_{\mathbf{k},\sigma}$ is the usual normal ordered FeSe number operator. Since the e-ph interaction is purely intraband we can treat each band individually and the band index i has been suppressed for clarity.

The Hamiltonian (6) can be solved exactly. The eigenstates and eigenenergies are given by

$$\begin{aligned} |\{n_{\mathbf{k}\sigma}\}, n\rangle &= \frac{e^{-a_c^2/2}}{\sqrt{n!}} (a^+ + \delta\hat{N} a_c)^n e^{-\delta\hat{N} a_c a^+} |\{n_{\mathbf{k}\sigma}\}, 0\rangle \\ E_{\{n_{\mathbf{k}\sigma}\},n} &= \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma} n_{\mathbf{k}\sigma} + n\hbar\Omega_0 - \frac{g^2}{\hbar\Omega_0} \delta N^2 \end{aligned} \tag{7}$$

where $\{n_{\mathbf{k}\sigma}\}$ are the occupations of the FeSe electrons, n is the number of phonon quanta, and $|\{n_{\mathbf{k}\sigma}\}, 0\rangle$ denotes the electron configuration and phonon vacuum. The key parameter in Eq. (7) is $a_c = g/\hbar\Omega_0$, the dimensionless e-ph coupling strength. The last term in Eq. (7) is the phonon-mediated electron-electron attraction $-\frac{g^2}{\hbar\Omega_0}\delta N^2 = -v_{\text{eff}}\delta N^2$, which has a strength

$$v_{\text{eff}} = \hbar\Omega_0 a_c^2. \tag{8}$$

Given Eq. (7), it can be shown straightforwardly that the ratio between the ARPES intensity of the nth replica band and the main band is given by

$$\frac{I_n}{I_0} = \frac{a_c^{2n}}{n!}. \tag{9}$$

Therefore by measuring the intensity ratio between the first replica band the main band we can get an estimate of a_c^2. Once a_c^2 is known we can estimate the strength of phonon-mediated attraction using Eq. (8).

Since for the Fermi-level crossing electron band near m our experimental results places the following lower bound on the intensity ratio

$$\frac{\text{Intensity of the first replica band}}{\text{Intensity of the main band}} \geq \frac{1}{6.2} \tag{10}$$

we deduce the dimensionless e-ph coupling constant to be $a_c \approx 0.4$ and $v_{\text{eff}} \approx 0.16\hbar\Omega_0$. If we use $\hbar\Omega_0 = 80$ meV (see later) a phonon-mediated attraction strength $v_{\text{eff}} \approx 13$ meV is obtained.

6 A more realistic model for the replica band

In this section we consider a multiband system where the i-th band is assigned a bare dispersion $\epsilon_i(\mathbf{k})$, which coupled to a dispersionless phonon band of frequency Ω_0. For simplicity we further assume that the coupling is purely intraband in the vicinity of $\mathbf{q} = 0$. The generic form of the Hamiltonian reads as

$$H = \sum_{\mathbf{k},i,\sigma} \epsilon_i(\mathbf{k}) c^\dagger_{i,\mathbf{k},\sigma} c_{i,\mathbf{k},\sigma} + \hbar\Omega_0 \sum_{\mathbf{q}} (b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + 1/2) + \frac{1}{\sqrt{N}} \sum_{\mathbf{k},\mathbf{q},i,\sigma} g(\mathbf{q}) c^\dagger_{i,\mathbf{k}+\mathbf{q},\sigma} c_{i,\mathbf{k},\sigma} (b^\dagger_{\mathbf{q}} + b_{-\mathbf{q}}). \tag{11}$$

Here, $c^\dagger_{i,\mathbf{k},\sigma}$ ($c_{i,\mathbf{k},\sigma}$) creates (annihilates) an electron of spin σ in band i with momentum $\mathbf{k}$, $b^\dagger_{\mathbf{q}}$ ($b_{\mathbf{q}}$) creates (annihilates) a phonon with momentum $\mathbf{q}$, and $g(\mathbf{q})$ is the momentum dependent e-ph coupling constant.

Given Eq. (11) we are now able to compute the electron spectral function for each band $A_i(\mathbf{k},\omega) = -2\text{Im}[G_i(\mathbf{k},\omega)]/\pi$, where $G_i^{-1}(\mathbf{k},\omega) = \omega - \epsilon_i(\mathbf{k}) - \Sigma_i(\mathbf{k},\omega)$ is the dressed Greens function and $\Sigma_i(\mathbf{k},\omega)$ is the self-energy due to the e-ph interaction which we calculate self-consistently using the Eliashberg formalism.[34] More explicitly we solve the real-axis Eliashberg equations in the normal state while retaining the full momentum dependence of the band structure and e-ph coupling constant.

Motivated by the discussion in section 4 we adapt the following $g(\mathbf{q})$

$$g(\mathbf{q}) = g_0 \exp(-|\mathbf{q}|/q_0), \tag{12}$$

where g_0 sets the overall strength of the interaction. The self-energy for each band $\Sigma_i(\mathbf{k}, \omega + i\delta)$ is found by iteratively solving[35,36]

$$\begin{aligned}\Sigma_i(\mathbf{k}, \omega + i\delta) &= \frac{1}{N\beta}\sum_{m,\mathbf{p}} D(\mathbf{p}-\mathbf{k}, \omega - i\omega_m) G_i(\mathbf{p}, i\omega_m) \\ &+ \Bigg[\left[n_b(\Omega_0) + 1 - n_f(\omega - \Omega_0) \right] \frac{1}{N}\sum_{\mathbf{p}} |g(\mathbf{q})|^2 G_i(\mathbf{p}, \omega - \Omega_0) \\ &+ \left[n_b(\Omega_0) + n_f(\omega + \Omega_0) \right] \frac{1}{N}\sum_{\mathbf{p}} |g(\mathbf{q})|^2 G_i(\mathbf{p}, \omega + \Omega_0), \Bigg] \end{aligned} \tag{13}$$

where $\mathbf{q} = \mathbf{p} - \mathbf{k}$ is the momentum transfer, $\beta = 1/(k_B T)$ is the inverse temperature, n_b and n_f are the Bose and Fermi occupation numbers, respectively,

$$D(\mathbf{q}, x) = g^2(\mathbf{q}) \frac{2\Omega_0}{\Omega_0^2 - x^2}. \tag{14}$$

For simplicity, we have modeled the band dispersion in the $\Gamma - M$ direction with one-dimensional cosine bands $\epsilon_{e,h}(\mathbf{k}) = -2t_{e,h}\cos(k/a) - \mu_{e,h}$ where $t_e = 125$ meV, $t_h = -30$ meV, $\mu_e = -185$ meV, and $\mu_h = 175$ meV. The total ARPES intensity is then given by a sum of the spectral functions for each band, multiplied by a matrix element $|M_i|^2$,

$$A(\mathbf{k}, \omega) = \sum_i |M_i|^2 A_i(\mathbf{k}, \omega). \tag{15}$$

The relative intensities between the main electron and hole bands are set by the relationship between $|M_e|^2$ and $|M_h|^2$, while the intensities between the main bands and the replica bands are set by Ω_0, g_0, q_0. The parameter values which yielded the fit to the experimental data plotted in the main text are given by $\Omega_0 = 80$ meV, $g_0 = 0.04$ eV, and $q_0 = 0.3/a$ ($a = 3.9$ Å, with $|M_e|^2 = 4.3|M_h|^2$. These parameters correspond to a total dimensionless e-ph coupling strength

$$\lambda = \frac{2}{N\Omega_0} \frac{\sum_{\mathbf{k},\mathbf{q}} |g(\mathbf{q})|^2 \delta(\epsilon_{\mathbf{k}}) \delta(\epsilon_{\mathbf{p}-\mathbf{q}})}{\sum_{\mathbf{k}} \delta(\epsilon_{\mathbf{k}})} = 0.5. \tag{16}$$

Finally by integrating out the phonons in the full e-ph Hamiltonian Eq. (11) we arrive at an effective Hamiltonian (Eq. 21) with $v_{\text{eff}} = \frac{\pi g_0^2 q_0^2}{\hbar \Omega_0}$. For the parameters used in Fig. 4c of the

main text we arrive at $v_{\rm eff} \approx 9$ meV. Though this is comparable to the value obtained from the simpler model used in section 5, given the uncertainty in the determination of $v_{\rm eff}$, we adopt an order of magnitude value for $v_{\rm eff}$ = 10 meV, which is the value used in the main text. We note that the exponential $\mathbf{q}$-dependence of $g(\mathbf{q})$ in Eqn. (13), with an extremely small value of q_0 (0.3/a, or 5% of $2\pi/a$), means that the e-ph coupling is extremely focused in the forward direction. This is much sharper than the $1/q$ dependence expected from Coulomb screening, which usually is considered as an extreme case. The consequences of this for pairing in unconventional superconductors are discussed in the following section.

The effects of the q_0 on the replica band can be seen in the momentum distribution curves (MDCs) of the dispersion, plotted in ED Fig. 5. 5a shows the calculated MDCs of the main band and replica band, plotted at the same energy relative to their respective band bottoms. ED Fig. 5b plots a similar set of MDCs for the data. A momentum independent background has been subtracted from the replica MDC data. We find satisfactory agreement between the two, namely, the peaks of the replica bands are broadened relative to those of the main band. The momentum-dependent background, e.g. contribution from the hole band, is not removed. We believe this is the cause of the extra broadening in the data.

7 Phonon-mediated contributions to unconventional superconductivity

The strong forward scattering nature of the coupling to the $SrTiO_3$ mode is unusual. Such a $\mathbf{q}$-dependence will be attractive in most pairing channels, including those established by spin fluctuations, while in other cases, it will not degrade pairing as would be the case for phonons with an isotropic coupling.[8-14] This can be seen from some general considerations. The symmetry of the superconducting order parameter $\Delta_{\mathbf{k}}$ is determined by the sign and momentum

structure of the pairing interaction $V_{\mathbf{k},\mathbf{p}}$ via the BCS gap equation

$$\Delta_{\mathbf{k}} = -\sum_{\mathbf{k},\mathbf{p}} V_{\mathbf{k},\mathbf{p}} \frac{\Delta_{\mathbf{p}}}{2E_{\mathbf{p}}} \tanh\left(\frac{\beta E_{\mathbf{p}}}{2}\right), \tag{17}$$

where $E_{\mathbf{k}} = \sqrt{(\epsilon_{\mathbf{k}} - \mu)^2 + \Delta_{\mathbf{k}}^2}$. Here we consider a multi-channel mechanism for superconductivity, where pairing is mediated by the sum of a repulsive electronic mechanism and an attractive phonon mechanism. The effective interaction can be written as $V_{\mathbf{kp}} = V_{\mathbf{kp}}^{el} - V_{\mathbf{kp}}^{ph}$. In the absence of the phonons, the repulsive interaction often results in a gap symmetry where $\Delta(\mathbf{k})$ changes sign on the Fermi surface. However, the exact symmetry depends on several factors, including the Fermi surface topology. Several commonly realized $\Delta(\mathbf{k})$ that are relevant to this discussion are sketched in ED Fig. 6.

The precise role of the phonons in such a case will depend on a number of factors including the momentum dependence of their coupling constant $g(\mathbf{q})$, the topology of the Fermi surface, and the symmetry of the gap. From the general structure of the gap equation Eq. (17) one can see that an additional attractive interaction will increase $\Delta_{\mathbf{k}}$ further when $V_{\mathbf{k},\mathbf{p}}$ connects regions of the Fermi surface where $\Delta_{\mathbf{p}}$ and $\Delta_{\mathbf{k}}$ have the same sign.[8, 13, 14] Conversely, the same interaction will reduce $\Delta(\mathbf{k})$ when it connects regions of the Fermi surface where $\Delta_{\mathbf{k}}$ changes sign. For example, in the d-wave cuprates bond-buckling oxygen modes (which favor $\mathbf{q} \sim 0$ coupling) are pairing while the bond-stretching oxygen modes (which favor $\mathbf{q} = (\pi/a, \pi/a)$) are pair breaking.[10]

The total contribution to pairing is determined by the sum of all scattering processes, weighted by $V_{\mathbf{kp}}$. This can be parameterized by a projected coupling

$$\lambda_{\phi}^{ph} \propto \frac{\int d\mathbf{k} \int d\mathbf{p} \Delta^*(\mathbf{k}) V_{\mathbf{kp}}^{ph} \Delta(\mathbf{p})}{\int d\mathbf{k} \left|\Delta\right|^2 (\mathbf{k})}, \tag{18}$$

which will be > 0 for a mode which contributes overall to pairing, < 0 for a mode which degrades pairing, and $= 0$ for a mode which does not contribute in any way. The forward-focused interaction inferred here is a unique case as it results in values of $\lambda_{\phi}^{ph} \geq 0$. Several

scattering processes are illustrated ED Fig. 6 in this limit. From these pictures it is clear that in many cases a forwardly focused e-ph interaction will enhance $\Delta(\mathbf{k})$ and thus give an increase to T_c. For example, for the d-wave cuprates (6a) or the pnictides with an $s_\pm$ (6b) gap, the phase space for small $\mathbf{q}$ scattering with the same sign gap is larger than the phase space for a sign change. Thus $\lambda_\phi^{ph} > 0$ and forward scattering enhances T_c. The same holds true in the case of FeSe with two M-centered electron pockets and the same sign gap. In the worse case scenarios, such as those shown in Fig. 6d and 6e, the scattering will average to zero giving no detrimental contribution. These general considerations highlight the importance of the forward-focused electron-phonon coupling which could have broader implications. As will be seen in the next section, such forward-focused attractive interaction significantly increases pairing in the specific case of FeSe/STO.

8 Estimating the pairing temperature enhancement

We now turn to an estimate of the T_c enhancement. In the absence of the e-ph interaction we assume the following *effective* Hamiltonian[23]

$$H_{\text{eff}} = \sum_{\mathbf{k}}{}' \sum_{\sigma} \epsilon(\mathbf{k}) c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + 2 \sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j, \tag{19}$$

In Eq. (19) the primed sum $\sum_{\mathbf{k}}{}'$ denotes a sum within a thin shell around the Fermi surface, $c^\dagger_{\mathbf{k}\sigma}$ creates a spin σ electron with momentum $\mathbf{k}$ in the band eigenstate, and $\vec{S}_i$ is the spin of electron at site i. The form of J_{ij} is taken to be

$$\begin{aligned} J_{ij} &= J_1 \text{ for nearest neighbors i, j} \\ &= J_2 \text{ for next nearest neighbors i, j.} \end{aligned} \tag{20}$$

To isolate the role of the substrate phonon, we assume that pairing is caused by the magnetic exchange term in Eq. (19) in the absence of the e-ph interaction.

The theoretical band structure we use in the calculation below is shown in ED Fig. 7a.[22] Here the hole band near Γ is below the bottom of the electron band at M. Note that because our experiment shows two slightly split electron pockets (see ED Fig. 7) we turned on a small hybridization between the electron pockets. The calculated split pockets are shown in ED Fig. 7b.

The e-ph interaction introduces an effective attractive interaction. After integrating out the phonon degrees of freedom, the Hamiltonian $H_{\rm eff}$ is supplemented with a term

$$H_{\rm eff} \to H_{\rm eff} - \sum_{\mathbf{k},\mathbf{p},\mathbf{q}} \sum_{\sigma,\sigma'} \left(\frac{v_{\rm eff}}{2\pi q_0^2} e^{-|\mathbf{q}|/q_0} \right) c^\dagger_{\mathbf{k}+\mathbf{q},\sigma} c_{\mathbf{k}\sigma} c^\dagger_{\mathbf{p}-\mathbf{q}\sigma'} c_{\mathbf{p}\sigma'}. \tag{21}$$

After turning on the phonon-mediated attraction we calculate the ratio $\Gamma(v_{\rm eff})/\Gamma(v_{\rm eff} = 0)$ where Γ enters the equation for T_c via the BCS expression (note we are describing the Cooper pair forming, i.e., a mean-field, transition)

$$k_B T_c = 1.14\,(\text{ cutoff energy})\, e^{-\frac{1}{\Gamma(v_{\rm eff})}}. \tag{22}$$

Our ultimate goal is to make a rough estimate of the T_c enhancement. To this end we set the cutoff energy to 65 meV, i.e., the energy difference between the Fermi energy and the bottom of the electron band, and take $T_c = 40$ K in the absence of the e-ph interaction, where the transition temperature corresponds to an upper bound for current iron-based superconductors.[26] In addition to these parameters, an estimate of $J_{1,2}$ in Eq. (19) is needed. These are the quantities for which we have the least information. We therefore take a conservative approach and assume generous values for $J_{1,2}$. As such we overestimate the electronic contribution to pairing and ask how the e-ph interaction further enhances T_c. In this way we will tend to underestimate the effect of the e-ph interaction.

In the literature estimates of $J_{1,2}$ usually come from studying the spin excitations by neutron scattering, whose results are fitted to some kind of Heisenberg Hamiltonian. The results

are typically reported as SJ_{ij}, where S is the magnitude of the spin. In many cases the estimated J_1 are highly anisotropic, and can even change sign along orthogonal directions. In the following we shall take the maximum value of J_1 along the two orthogonal directions known to us and assume as isotropic antiferromagnetic J_1. As to the value of J_2 we shall treat J_2/J_1 as a parameter and set $J = \sqrt{J_1^2 + J_2^2} = 30$ meV.[26]

We first examine what happens as we vary the ratio of J_2/J_1. ED Fig. 8 plots the phase diagram of the $J_1 - J_2$ model in the absence of the electron-phonon interaction. In this diagram the ratio of J_2/J_1 controls the pairing symmetry. As the ratio is varied from 0 to $\tan(\pi/2)$, a symmetry change occurs at $J_2/J_1 \sim 0.31$ at which the pairing symmetry changes from d-wave to s-wave. Note that the $s_\pm$ pairing widely perceived to be the likely pairing symmetry for iron pnictides, which have hole pockets around the Brillouin zone center, is not found for the band structure relevant to the 1UC FeSe, where there are only two slightly separated electron pockets at the Brillouin zone corner and no pockets in the center. Three considerations suggest the physically relevant J_2/J_1 should fall in the s-wave region: 1) The observation of a full gap in the 1UC system as reported by several ARPES experiments.[3] 2) A full gap is observed in systems with similar Fermi surface topology, namely, $\mathrm{K_xFe_{2-y}Se_2}$.[37] 3) The observation of striped AF phase in $\mathrm{K_xFe_{2-y}Se_2}$ at compositions close to the superconducting phase.[38] In the $J_1 - J_2$ model the striped AF phase is only realized if $J_2 \gtrsim 0.5J_1$.

Because of these reasons, for calculating the T_c enhancement we limit the range of J_2/J_1 to include only values where the gap function is s-wave, i.e. the gap does not go to zero at any point in momentum space. In this entire pairing range, the small q-phonon reinforces the magnetic pairing and raises the pairing temperature. In the d-wave region the effects of the electron-phonon interaction on pairing are very weak, for the reasons given in section 7. Fig. 4d plots the T_c enhancement as a function of v_{eff}/J for several values of J_2/J_1. We use $q_0 = 0.1\pi/a$ in Eq. (21), which is close to the value obtained by modeling the spectral functions

in section 6 (i.e. $0.3/a$). The results are shown in Fig. 4d. Using a $v_{\rm eff}$ = 10 meV, we calculate $v_{\rm eff}/J_{\rm eff} = 0.3$. This yields a T_c enhancement of $\approx$ 1.5.

As discussed earlier the above numbers should be viewed as a conservative estimate of the T_c enhancement, for we have used the smallest intensity ratio between the first replica band and the main band, and we also used the largest effective antiferromagnetic exchange constant. Of course we understand predicting T_c is a very tricky task. The purpose of our estimate here is to raise a reasonable possibility that the electron-phonon interaction in question can significantly enhance the Cooper pairing temperature in single UC FeSe/STO.

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