Films of iron selenide (FeSe) one unit cell thick grown on strontium titanate (SrTiO$_3$ or STO) substrates have recently shown superconducting energy gaps opening at temperatures close to the boiling point of liquid nitrogen (77 kelvin), which is a record for the iron-based superconductors. The gap opening temperature usually sets the superconducting transition temperature $T_c$, as the gap signals the formation of Cooper pairs, the bound electron states responsible for superconductivity. To understand why Cooper pairs form at such high temperatures, we examine the role of the SrTiO$_3$ substrate. Here we report high-resolution angle-resolved photoemission spectroscopy results that reveal an unexpected characteristic of the single-unit-cell FeSe/SrTiO$_3$ system: shake-off bands suggesting the presence of bosonic modes, most probably oxygen optical phonons in SrTiO$_3$ (refs 5–7), which couple to the FeSe electrons with only a small momentum transfer. Such interfacial coupling assists superconductivity in most channels, including those mediated by spin fluctuations$^8$$^{–}$$^{14}$. Our calculations suggest that this coupling is responsible for raising the superconducting gap opening temperature in single-unit-cell FeSe/SrTiO$_3$.

The dramatic enhancement of the superconducting transition temperature $T_c$ in FeSe, from 8 K in bulk$^{11}$ to nearly 70 K when grown as a single-unit-cell (1UC) layer on SrTiO$_3$, has generated research interest because it suggests that Cooper pairing can be strengthened in non-bulk systems. We begin by studying the electronic structure of the 1UC FeSe/SrTiO$_3$ film. Growth details are discussed in the Supplementary Information and Extended Data Fig. 1. The electronic structure is plotted in Fig. 1a–f and is consistent with previous reports$^{12}$$^{–}$$^{14}$. The Fermi surface of the 1UC film consists of electron-like (convex up, labelled A) pockets centred around the Brillouin zone center (M-point) with a band bottom 60 meV below the Fermi energy ($E_F$). Further analysis reveals that there are two nearly degenerate electron bands at M (see Supplementary Information). Below the bottom of A, one hole-like (concave down, labelled B) band is clearly resolved. The zone centre (I) consists of another hole-like band (labelled D) with a top located 80 meV below $E_F$. The temperature evolution of the band structure is shown in Fig. 1a–f and shows typical superconducting gap behaviour, with the electron band bending away from the Fermi energy (backbending) at the Fermi momentum $k_F$. The symmetrized energy distribution curves (EDCs) shown in Fig. 2c are fitted using a phenomenological model (ref. 16) to reveal a 13-meV gap plotted in Fig. 2d. Using a mean-field formula we obtain $T_c = 58 \pm 7$ K, consistent with previous results within experimental uncertainty$^{12}$$^{–}$$^{14}$. Analysing the energy gap at different points (Fig. 2e and f), we observe a round, un-nested Fermi surface with a nearly uniform gap, making it unlikely that the gap is caused by other instabilities such as charge density waves.

We now turn to the most unexpected aspect of our 1UC data: the clearly resolved replica bands labelled A’ and B’ in Fig. 1d–f. Aside from an approximately 100-meV relative shift in energy, all features of A are replicated in A’. This includes the backbending near $k_F$, seen in Fig. 3. Similarly B and B’ are also separated by the same energy shift with nearly the same dispersion. In Fig. 1d we mark the peaks associated with A, A’ and B, B’ blue and red, respectively, to illustrate their presence even in the raw EDC data. In the Supplementary Information we rule out the possibility that such replicas could be caused by band structure effects (such as quantum well states$^{17}$$^{–}$$^{18}$) and further discuss weaker features such as C and D.’

We believe the replica bands are due to the shaking off of quanta of the bosonic modes in SrTiO$_3$, reminiscent of the vibron shake-offs in the photoemission spectra of H$_2$ molecules$^{19}$. In the Supplementary Information we make the case that such bosonic modes are associated with a high-energy SrTiO$_3$ oxygen phonon band$^{5}$$^{–}$$^{6}$ near 100 meV, although its exact energy may be modified somewhat by the presence of the overlaid film. This identification is supported by recent high-resolution angle-resolved photoemission spectroscopy (ARPES) on SrTiO$_3$ surface states, which shows a phonon-induced hump at approximately 100 meV away from the main band$^{20}$$^{–}$$^{21}$ and through inelastic neutron scattering. Resolving replicas of an entire band with such a clear dispersion, as seen in Fig. 1d, is unprecedented in a solid. This is made possible here by the substantial electron–phonon coupling, the fact that the collective mode energy is greater than the width of the electron band below $E_F$, and most importantly, the fact that the electron–phonon coupling allows only a small momentum transfer to the electron (see below and Supplementary Information).

We now turn to the multi-unit-cell films. We have measured 2UC and 30UC FeSe films grown on similar Nb-doped SrTiO$_3$ substrates. In Fig. 3a–f we compare the second energy derivative of the ARPES spectra for the 1UC film (Fig. 3a and d), 2UC (Fig. 3b and e) and 30UC (Fig. 3c and f) films. We find that the 2UC and 30UC band structure consists of both electron-like and hole-like bands crossing $E_F$ near M, similar to what has been observed for bulk FeSe$^{21}$. This is very different from the 1UC film’s band structure, where only electron bands cross $E_F$. This implies that the 1UC film is much more heavily electron-doped than even the 2UC film$^4$. Most importantly, we observe neither a superconducting-like energy gap nor replica bands in the multi-UC films. A complete comparison of the band structures can be found in Extended Data Fig. 2.

The temperature evolution of the replica bands in the 1UC film is plotted in Fig. 3g–j (additional temperatures are plotted in Extended Data Fig. 3). We see that these replica bands, and hence this anomalous electron–phonon coupling, persist to temperatures significantly above the gap-opening temperature. The contrast between the 1UC and multi-UC
Figure 1 | Fermi surface map and high symmetry cuts of 1UC FeSe on SrTiO₃. a, Plot of the Fermi surface with only electron pockets located at the zone corner (M-point; kₓ and kᵧ refer to momentum directions in the Brillouin zone). All momentum axes in this paper are given in units of inverse angstroms relative to the high-symmetry points. Red dots are the approximate points on the Fermi surface where gaps were extracted in Fig. 2e, f. b, c, High-symmetry cuts along the purple line plotted in a, taken at 16 K. b and c are centred at Γ (zone centre) and M, respectively (kₚₜₐᵣ refers to momentum along the C–M direction). The hole band seen in b is located 80 meV below E_F. In c a different colour scale highlights two important features: the electron band with a minimum at 60 meV below E_F (labelled A), and a replica electron band (labelled A’), which is located 100 meV below the former and sits on top of a broad hole band. d, EDCs at M shown as a waterfall plot, with markers indicating band peaks. e, f, Second derivatives in energy of the high-symmetry cuts from b and c. An additional weaker replica, labelled C, can now be seen at M in f, sitting below A, and at the Γ point in e we see the hole band and a corresponding replica, labelled D and D’, respectively. Data for all figures were obtained using samples grown with the same method, as described in the Supplementary Information. Spectra shown in all figures are representative data taken from six different samples that yielded comparable results.

Figure 2 | Temperature dependence of the 1UC film energy gap at the M-point. a, b, Plots of the Γ–M high-symmetry dispersion EDCs at 16 K and 64 K respectively, with the Fermi–Dirac distribution factored out. The cut direction is given by the green arrow marked in Fig. 1c. The red trace represents the EDC at the Fermi momentum kₓ. c, Plot of the evolution of the symmetrized EDCs at kₓ as a function of temperature, where we observe a gap closing between 50 K and 64 K. d, The gap evolution as a function of temperature, with gap magnitude Δ extracted using the model in ref. 16. Error bars in energy include the drift of E_F as measured relative to a gold reference. Error bars in temperature include temperature differences within the sample manipulator. A fit to a mean-field type of order parameter is plotted, giving a gap closing temperature of 58 ± 7 K, with the range representing the uncertainty in fitting parameters. e, Symmetrized EDCs at different kₓ values along the Fermi surface at 16 K, in the direction indicated by the red arrow in Fig. 1a. f, Polar plot of the gap of the EDCs from e. Error bars again include the drift of E_F as measured relative to a gold reference.
films suggests that the SrTiO$_3$ phonon which causes the replica bands is also responsible for enhancing Cooper pairing. The fact that the replica bands in the 1UC film follow the dispersion of the main band so closely suggests that, upon either absorption or emission, the phonons can transmit only small momenta to the electron. Such a strong forward-focused electron–phonon interaction can enhance Cooper pairing in most symmetry channels, including those with a sign change (see Supplementary Information and Extended Data Fig. 6).

To extract the strength of the electron–phonon coupling, we perform a high-statistics scan at M at low temperature, plotted in Fig. 4a. Using a spline background, we find a lower bound of 1/6 for the intensity ratio of the replica band to the main band (see Fig. 4b and Extended Data Fig. 4). We then take the intensity ratio as input and theoretically estimate the electron–phonon coupling strength (see Supplementary Information). Plotted in Fig. 4c is a simulated spectral function calculated using a model in which both the electron and hole bands couple to a flat phonon band with energy 80 meV. By tuning the coupling strength and the forward–focusing parameter we can reproduce a band–replica separation of approximately 100 meV. The simulated EDC is plotted in Fig. 4b. The ARPES spectrum is well reproduced, especially the abrupt loss in spectral weight of band A’ beyond a certain momentum window, and the momentum broadening of the bands, plotted in Extended Data Fig. 5.

From the intensity ratios we obtain the electron–phonon coupling constant $\lambda \approx 0.5$, which is substantial considering that only a narrow range of phonon modes at such high frequency contribute to the coupling (owing to nearly forward scattering). This estimate in turn yields an effective phonon-mediated attraction strength $v_{\text{eff}} \approx 10$ meV (see Supplementary Information). Under the assumption that Cooper pairing is

![Figure 3](image1.png) Dependence of electronic structure on FeSe film thickness. a, Spectrum of the 1UC film at Γ. b, Spectrum of the 2UC film. c, Spectrum of the 30UC film. d, Spectrum of the 1UC film at M. e, M spectrum for the 2UC film. f, M spectrum for the 30UC film. Data in a and f have been symmetrized around Γ and M, respectively, as indicated by the green line, while data on the 1UC and 2UC films are unsymmetrized. The colour-coded lines in a and d are guides to the eye, with solid lines denoting the main band and dashed lines corresponding to the replicas. The main bands and replicas are colour-coded according to Fig. 1. We do not observe replica bands for either the 2UC and 30UC films. Raw spectra can be found in Extended Data Fig. 2.

![Figure 4](image2.png) Extraction of the electron-phonon coupling and determination of $T_c$ enhancement. a, High-statistics scan at M taken at low temperature (10 K). The spectral weight is integrated over the momentum range indicated by the red rectangle to obtain better statistics for a single EDC. b, The integrated EDC at M (after background subtraction) and the EDC from our calculations. Peaks corresponding to the bands are labelled according to Fig. 1a. c, Model spectral function calculation including both hole and electron bands coupled to a dispersionless 80 meV phonon mode (see Supplementary Information for details). The black dotted line indicates the EDC plotted in b. d, Plot of $T_c$ enhancement as a function of effective attractive electron–electron interaction strength $v_{\text{eff}}/J$. Shown are plots for three different values of $J_f/J_c$. The parameters we used to construct this curve (see Supplementary Information for additional details) are $q_0 = 0.1\pi/a$ ($a = 3.9 \text{ Å}$), $J_c = 30$ meV, cutoff energy $= 65$ meV, and $T_c = 40$ K in the absence of electron–phonon interaction. Using the extracted parameters, we place the lower bound of the enhancement factor at 1.5.
caused by the magnetic interaction in the absence of the electron–phonon interaction, and following the effective Hamiltonian approach described in ref. 23, we first determine the pairing symmetry without the electron–phonon interaction as a function of the ratio between the nearest- and second-neighbour magnetic exchange constants $J_1$ and $J_2$. The band structure we use is plotted in Extended Data Fig. 7 (ref. 22). We note that for the physically relevant $J_2/J_1$ ratio (>0.5) the pairing is an in-phase $s$-wave between the two electron pockets $^{24,25}$; see Extended Data Fig. 8. We then determine the enhancement of the Cooper pairing temperature as a function of the ratio between $v_{\text{eff}}$ and the antiferromagnetic exchange constant $J = \sqrt{(J_1^2+J_2^2)}$ (see Supplementary Information). We obtain $J = 30$ meV from the largest nearest- and second-neighbour exchange constants determined from neutron scattering experiments known to us $^{26}$, thus overestimating the magnetic coupling. Figure 4d plots the $T_c$ enhancement as a function of $v_{\text{eff}}/J$ for varying ratios of $J_1$ to $J_2$. Using the $v_{\text{eff}}$ extracted from our data and the above estimate of $J$, we determine the enhancement factor to be about 1.5. This enhancement is a lower bound, because we use the most conservative estimate of the electron–phonon coupling, and the largest $J$ value. For bulk materials with similar band structures, for example, $\text{K}_2\text{Fe}_{2-y}\text{Se}_2$ (ref. 27), or materials from which we obtain the above stated $J$ value$^{26}$, the $T_c$ values range from 30 K to 40 K. Multiplying by the enhancement factor yields a gap-opening temperature in fairly good agreement with our films. It should be noted that we do not use the $T_c$ of bulk FeSe because our films are heavily electron doped and hence have a very different band structure from that of bulk FeSe.

Motivated by the physical picture presented here, we propose a heterostructure where $\text{UC}_2\text{Fe}_3\text{Se}_8$ is sandwiched on both sides, effectively doubling $v_{\text{eff}}$. A simple reading of Fig. 4d suggests a $T_c$ enhancement of around 2.5, placing the Cooper pairing temperature well above the temperature of liquid nitrogen.

Online Content Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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Author Information Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to Z.-X.S. (zxshen@stanford.edu).
Extended Data Figure 1 | Reflection high-energy electron diffraction (RHEED) images observed during FeSe growth. 

a, RHEED image of SrTiO$_3$ substrate after degassing at 450 °C for 1 h. Red box highlights the region integrated for monitoring RHEED oscillations. 
b, Surface reconstruction as observed by RHEED at annealing temperatures. 
c, RHEED image of FeSe 1UC film showing uniform streaks typical of an atomically flat thin film. 
d, RHEED intensity for integration region shown in a (black). The second derivative of the intensity curve (red), with numbers indicating the number of layers grown highlights the RHEED oscillations signalling the completion of a unit cell after about 30 s.
Extended Data Figure 2 | Raw spectra and second derivatives of 1UC, 1.7UC, 2UC and 30UC films. a, f, k, p. Plots of the raw spectrum at Γ, the second derivative at Γ, the raw spectrum at M, and the second derivative at M for the 1UC film, respectively, taken at 10 K. b, g, l, q. The same plots for the 1.7UC film taken at 13 K. c, h, m, r. The same plots for the 2UC film taken at 15 K. d, i, n, s. The same plots for the 30UC film taken at 50 K. e, j, o, t. The same plots for the 30UC film taken at 140 K. Data for the 30UC film are symmetrized about the high-symmetry points (indicated by the green line), and were taken with 25-eV photons. We observe a band splitting in the 30UC film at M, at low temperature (s). This band splitting closes at higher temperature (140 K), where we now observe only one band (t).
Extended Data Figure 3 | Temperature evolution of the M point spectrum of the 1UC film. a, Spectrum at 10 K, where we can clearly see the backbending of the replica bands, exactly like the main bands near $E_F$. b, Spectrum at 30 K. c, Spectrum at 50 K. d, Spectrum at 70 K. e, Spectrum at 90 K. f, Spectrum at 120 K. The replica bands persist up to temperatures significantly higher than the gap-opening temperature.
Extended Data Figure 4 | Fitting the intensities of the ARPES spectra at M.

a, Plot showing two different backgrounds used in the fitting. Using the blue circles as fixed points, we first modelled the background using a spline interpolation, plotted in red. The second fit used a Shirley background (see Supplementary Information), plotted in purple. b, Data and fitting with the spline background subtracted. We fitted to four Gaussian peaks, which are plotted separately for clarity. c, Data and fitting with the Shirley background subtracted. We restrict our fitting energy window to be from −0.32 eV to 0.03 eV.
Extended Data Figure 5 | Momentum distribution comparison between the main band and replica band. **a**, The momentum distribution curves (MDCs) of our theoretical calculation of the main electron band and the replica electron band with normalized intensities. The MDCs of both bands are taken at the same energies with respect to their band bottoms (see inset). The replica band peaks are broadened due to the electron–phonon coupling. **b**, The MDCs of our data, with a momentum-independent background subtracted from the replica band MDC. The momentum-dependent background—such as contributions from the hole band—is the likely cause of the extra broadening in the data.
Extended Data Figure 6 | Effects of electron-phonon coupling on different gap symmetries and Fermi surfaces. Cartoon sketches of the various Fermi surfaces and gap symmetries found or proposed in unconventional superconductors. $\lambda_{\text{ph}}$ is the projected coupling defined by equation (18) in the Supplementary Information. 

- **a.** Sketch for the copper oxides with a $d$-wave gap. 
- **b-e.** Various scenarios for the iron-based superconductors. Only one-quarter of the first Brillouin zone is shown for clarity. The thick blue and red lines indicate the phase of the gap. The arrows show various forward-focused scattering processes. The black arrows indicate scattering processes that connect portions of the Fermi surface with the same sign gap and are therefore pair-enhancing. The green arrows show pair-breaking processes which connect regions of the Fermi surface with different signs.
Extended Data Figure 7 | Input electronic structure for calculated $T_c$ enhancement. a, Calculated band structure used in our determination of $T_c$ enhancement. b, Calculated Fermi surface showing slightly split electron pockets. c, Dispersion along the M-point showing two nearly degenerate bands indicated with red and blue arrows. d, Momentum distribution curve (MDC) at $E_F$ showing peaks from the two bands plotted in c.
Extended Data Figure 8 | Phase diagram of the $J_1 - J_2$ model. The blue line represents $\sqrt{J_2^2 + J_2^2} = 1$. The red line represents the transition between different gap symmetries at $J_2/J_1 = 0.31$. Above the transition one finds a gap with $s$-wave symmetry. Below the transition one finds a gap with $d$-wave symmetry. Diagrams of the two different possible symmetries are drawn in their respective region of the phase diagram as insets. The lengths of the tick lines in the inset diagrams represent the magnitude of the gap, while the colour represents the sign (red for minus, blue for plus). The two electron pockets in the figure are separated for clarity.