Supplementary Information for
Emerging 2D-ferromagnetism and strong spin-orbit coupling at the surface of valence-fluctuating EuIr$_2$Si$_2$

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SUPPLEMENTARY NOTE 1

Assuming comparable photoionization cross sections for the $4f^6$ and $4f^7$ configuration at 145 eV photon energy [1], we expect for a mean valence of 2.8 at 7 K an intensity ratio $I^{3+}/I^{2+} \approx 10/3$ while $I^{3+}/I^{2+} \approx 1/3$ is observed experimentally. Taking a mean free path of the photoelectrons of 6 Å at 140 eV kinetic energy corresponding to about half of the lattice constant $d$ of the compound, the experimental result is in agreement with the assumption of a divalent fourth subsurface layer on top of the mixed valent bulk. At 200 K $I^{3+}/I^{2+} \approx 5/6$ is expected for a mean valence of 2.5 while the experimental value is only of the order 1/15. Thus, at 200 K also deeper lying Eu layers change obviously their valence towards lower values.

SUPPLEMENTARY FIGURE S1

Supplementary Figure S1. ARPES data taken from the Si-terminated surface of EuIr$_2$Si$_2$ at temperatures of 200 K, 100 K, 35 K and 7 K. The changes of the bulk Fermi surface sheet called Doughnut are compared with those derived from ab initio DFT calculations shown in the lower panel. The value for the valence $\nu$ used in the calculations was taken from the curve of the valence change shown in the inset of figure 1 of the manuscript. The violet dotted line is a guide to the eye to render the change in size of the Doughnut sheet of the bulk Fermi surface visible.
**SUPPLEMENTARY FIGURE S2**

Supplementary Figure S2. Valence changes derived from bulk sensitive XAS measurements (blue curve) and the size of the Doughnut Fermi surface sheet derived from *ab initio* DFT calculations (red curve). For the size the maximal expansion of the Doughnut in $k_x$ direction is taken and normalized to the length of the respective reciprocal lattice vector.

**SUPPLEMENTARY NOTE 2**

To determine the temperature $T_C$ at which the 4f moments in the subsurface Eu layer order we trace the evolution of the surface state in terms of its band dispersion in $\overline{X} - \overline{M}$ direction of the SBZ with temperature. In doing so we cooled the sample down and chose the direction in which the dispersion is asymmetric with respect to $\overline{M}$ (cf. Supplementary Figure S3(b)). Starting from 7 K, up to 40 K we increased the temperature stepwise by $\Delta T = 5$ K. Approaching the region of the magnetic transition we reduced the stepsize to $\Delta T = 1$ K until we reached $T = 54$ K which is well above the transition temperature and therefore characterized by a symmetric band dispersion (cf. Supplementary Figure S3(a)).

To extract $T_C$ we evaluate the surface state in terms of momentum distribution curves (MDCs) as it is indicated by white bars in the band maps given in Supplementary Figures S3(a) and S3(b). For both the left and right branch of the electron like surface band we present the temperature evolution of the MDCs separately in Supplementary Figures S3(c) and S3(d), respectively. Above the ordering temperature the band is subject to a symmetric Rashba splitting which is reflected
Supplementary Figure S3. Determination of the onset of magnetic order in the subsurface. ARPES band maps of the $\overline{M}$ surface state taken at (a) 53 K and (b) 7 K for the $\overline{X} - \overline{M}$ direction in which the asymmetry evolves at low temperatures. White bars indicate the MDCs that were traced with temperature to extract the magnetic transition temperature. The temperature evolution of the MDCs is shown for (c) the left and (d) the right branch of the electron like band. A drop in intensity and a change in peak separation observable at 48 K indicates the onset of magnetic order. The depicted momentum axis in (c) and (d) was derived from the ARPES data taken at 59 K. All other MDCs were shifted to align the peak at 0.53 Å$^{-1}$ and at 0.96 Å$^{-1}$, respectively.

in a double peak structure in the MDCs with an identical peak separation in both branches. For analysis the high-intensity peaks of the single MDCs were aligned with the one measured at 59 K, which serves as a reference. Thus, since the MCDs were slightly shifted the $k_\parallel$-axis refers only to the MDC measured at 59 K.

We start with the discussion of Supplementary Figure S3(c), that is the MDCs representing the left branch. With decreasing temperature the MDCs do not change fundamentally down to 49 K. At 48 K for the right, low-intensity peak a sudden drop in intensity is observable. Further lowering the temperature the peak moves to the left, merges with the high intensity peak into a single
peak and reappears as a shoulder on the left of the latter at lowest temperatures. As can be seen in Supplementary Figure S3(d) also the MDCs associated with the right branch of the band remain unaffected down to 49 K. At 48 K the left, low-intensity peak begins to move leftwards increasing the distance between the two peaks. The band splitting keeps increasing down to 10 K. In summary, our analysis points out that the magnetic transition in the subsurface takes place at 48 K.

SUPPLEMENTARY NOTE 3: X-RAY MAGNETIC CIRCULAR AND LINEAR DICHROISM

In order to confirm the magnetic properties at the surface of EuIr$_2$Si$_2$ we performed additional soft X-ray absorption measurements. In particular, we measured the magnetic circular and linear dichroism at the Eu $M_{4,5}$ edges using total electron yield detection which contains a sizable contribution from Eu located in the surface region [2–4].

The X-ray absorption measurements were done at the ESRF beamline ID32 using the high-field magnet end station [5, 6]. The samples were cleaved inside the high-field magnet at low temperature and a base pressure better than $1 \times 10^{-10}$ mbar. Measurements were done in normal incidence with the $c$ axis parallel to the incoming beam and at grazing incidence with an angle of 70° between the $c$ axis and the incoming beam. The $a$ axis was oriented to be in the horizontal plane, i.e. parallel to the $E$ vector of the X-rays for linear horizontally polarized light and normal incidence geometry. The light polarisation is always close to 100% for both linear and circular polarisation. Bulk magnetisation measurements were performed at $T = 5$ K in fields up to 7 Tesla using a SQUID-VSM (MPMS3, Quantum Design).

For the X-ray magnetic circular dichroism (XMCD) measurements a magnetic field was applied along the beam direction $\hat{u}_k$, i.e. out of surface plane. The XMCD signal, i.e. the difference in absorption for circular left and right polarisation, is proportional to the aligned moment along $\hat{u}_k$. The integrated XMCD signal of EuIr$_2$Si$_2$ as a function of applied magnetic field and temperature is shown in Figure S4 (right panel). For reference we also show the magnetisation curve obtained from bulk susceptibility measurements at $T = 5$ K. Another useful reference is the XMCD signal obtained from trivalent EuCo$_2$Si$_2$ which at low temperatures contains only a small contribution from van Vleck type bulk paramagnetism [7] and is dominated by the paramagnetic response of the Eu$^{2+}$ terminated surface. The field induced magnetic polarisation in EuIr$_2$Si$_2$ found from XMCD is larger than what is seen in bulk magnetisation measurements. Evidently, it has a second
**Supplementary Figure S4.** Left top: Experimentally observed XMCD in EuIr$_2$Si$_2$ as a function of applied magnetic field in normal and grazing incidence at $T = 5 \text{ K}$. The observation of a finite zero field XMCD signal at grazing incidence requires a net magnetic moment in the $ab$ plane which is not observed in the bulk. It thus confirms ferromagnetic in-plane order at the surface of this material. Left bottom: Full multiplet calculations of the XMCD of free Eu$^{2+}$ and Eu$^{3+}$ ions at $B = 9 \text{ T}$ at $T = 5 \text{ K}$. Right: Integrals over the absolute value of the XMCD signals of EuIr$_2$Si$_2$ at normal and grazing incidence. For reference, we also show the XMCD data for trivalent EuCo$_2$Si$_2$ during the same experiment and the previously reported low temperature bulk magnetisation of the two compounds for $B \parallel c$ [7, 8].

The contribution which can be easily polarised and for which saturation is reached already at a few Tesla. The comparison with the EuCo$_2$Si$_2$ XMCD shows that this contribution is not due to the Eu$^{2+}$ terminated surface. Hence, it is straightforward to assume that the additional magnetic contribution in EuIr$_2$Si$_2$ that saturates at a few Tesla already is coming from an easily polarised Si terminated Eu layer, i.e. the same surface that we studied with ARPES.

We get a direct evidence of ferromagnetic order in the Si-Ir-Si-Eu (SISE) surface block by looking at the zero field XMCD in grazing incidence geometry where it is sensitive to the in-plane net magnetic moment. The data is also shown in Fig. S4 and has been obtained from a freshly cleaved sample before applying a magnetic field. Already at zero field there is a sizable XMCD
Supplementary Figure S5. **Top:** Experimentally observed XMLD signal in EuIr$_2$Si$_2$ at $T = 5$ K and normal incidence. The finite XMLD signal in zero applied field requires the presence of in-plane magnetic order with moments along or close to the (100) direction. **Bottom:** Full multiplet calculations of the XMLD of Eu$^{2+}$ and Eu$^{3+}$ ions at $T = 5$ K with an exchange field of $\mu_B H_{ex} = 0.2$ meV applied along $a$.

signal in grazing incidence geometry which has the same shape as the high field XMCD obtained at normal incidence. This spectral shape is also well reproduced in full multiplet calculations [9] for a Eu$^{2+}$ ion at $T = 5$ K and $B = 9$ T applied along $c$ (Fig. S4, lower panel). The observation of a zero field XMCD signal in grazing incidence but not in normal incidence geometry shows that there must be an in-plane net magnetic moment with no or only a very small component along $c$. It therefore confirms the in-plane ferromagnetic order concluded from our ARPES results.

In addition to the XMCD measurements we also performed X-ray linear dichroism (XLD) measurements at normal incidence using the same experimental end-station. The alignment of the samples was the same as for the XMCD measurements, i.e. $a$ axis was in the horizontal plane and the $b$ axis in the vertical plane. At normal incidence, the $k$ vector of the incident light is parallel to the $c$ axis and a field of up to 4 Tesla can be applied along $a$. The XLD is the difference in the absorption of X-rays with linear horizontal or vertical polarisation, i.e. for the $E$ vector parallel to $a$ or $b$. In the tetragonal $I4/mmm$ structure of EuIr$_2$Si$_2$ the local point symmetry of the Eu ions is $D_{4h}$ and the $a$ and $b$ direction are exactly equivalent. Therefore, in the chosen experimental
geometry a crystal electric field (CEF) cannot lead to an XLD signal. However, as we show in Fig. S5 a small but clear XLD signal is observed for EuIr$_2$Si$_2$ already in zero magnetic field. We can exclude that this signal is due to sample misalignment, because the Eu$^{2+}$ ($L = 0, J = S = 7/2$) and the low temperature Eu$^{3+}$ ($J = 0$) electronic configurations with $2J + 1$ degeneracy are both not split by a CEF and therefore can also not produce an XLD even between the nonequivalent $a$ and the $c$ axis. By contrast, in the presence of magnetic order the $J = 7/2$ ground state of Eu$^{2+}$ will Zeeman-split due to the presence of the exchange field and give rise to X-ray magnetic linear dichroism (XMLD) which is what we see in the experiment. The shape of the experimental XMLD spectrum can be compared again with results of full atomic multiplet calculations which we show in the bottom panel of Fig. S5. The calculations have been performed for $T = 5$ K and assuming an exchange field $\mu_B H_{ex} = 0.2$ meV along the $a$ direction. Like for the XMCD we obtain a very good agreement between the calculated and experimentally observed dichroism. The experimental data also shows that the application of a very small magnetic field along the $a$ direction, $B = 30$ mT, strongly enhances the XMLD as one would expect as a consequence of the alignment of magnetic domains in the external field. We additionally observe a complete suppression of the XMLD signal when we apply $B = 5$ T along the $c$ direction (not shown here) which again is in line with expectations. We have seen that at $B = 5$ T along $c$ the subsurface XMCD signal has reached saturation and the ferromagnetically ordered moments will be fully aligned along $c$. As a consequence, in-plane XMLD should not be observed anymore.

In summary, both the observation of the XMCD as well as the XMLD in zero magnetic field prove the presence of in-plane magnetic order in the subsurface region. In addition to that we obtained complementary information from both techniques. The XMCD is proportional to $M \cdot \hat{u}_k$ averaged over all probed Eu sites. The zero field signal at grazing incidence but not at normal incidence therefore requires the presence of ferromagnetism perpendicular to the $c$ axis, i.e. in the plane. But it cannot tell, whether the moments are aligned along the (100) or the (110) direction because in both cases one obtains a finite projection of $M$ on $\hat{u}_k$. The XMLD in our geometry on the other hand is proportional to $(M \cdot \hat{u}_a - M \cdot \hat{u}_b)^2$ averaged over all sites. A zero field signal therefore does show the presence of in-plane order but cannot tell whether it is ferro- or antiferromagnetic. By contrast, it does contain more information about the orientation of the ordered moments in the plane, because moments along (110) would give the same projection of $M$ on the $a$ and $b$ axis and produce no XMLD, while moments along (100) give a finite XMLD. In combination, both techniques confirm the presence of in-plane ferromagnetic order in the subsurface Eu layer with moments aligned along the (100) direction, which is in line with the presented ARPES
Supplementary Figure S6. Band structure of the Si-terminated surface of the paramagnetic EuIr$_2$Si$_2$.  (a) Spin-resolved LDA band structure of the bulk-truncated symmetric 31-layer slab by the ELAPW method (see Supplementary Note 4) shown by fat bands along the $\bar{X} - \bar{M} - X$ line highlighted blue in graph (b). Blue and red fat bands show $S_y > 0$ and $S_y < 0$ states, respectively.  (b) The surface Brillouin zone showing the $\bar{X} - \bar{M} - X$ line, the Fermi-contour region of $\mathbf{k}$-space around $\bar{M}$, and the color code for the spin $x$- and $y$-projections.  (c) Spin-resolved LDA Fermi contours for the surface states.  (d) Spin-resolved band structure by the $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian (see Supplementary Note 4) for the momentum $\mathbf{k}$ within the vicinity of $\bar{M}$ on the $\bar{X} - \bar{M} - X$ line [see the vertical dashed lines in graph (a)].

Effective $\mathbf{k} \cdot \mathbf{p}$ model from first principles

In order to microscopically derive the effective $\mathbf{k} \cdot \mathbf{p}$ model for the considered surface states within the methodology of Ref. [10], we start with \textit{ab initio} calculations for the Si-terminated (001) surface.
of the paramagnetic EuIr$_2$Si$_2$, which we simulate by a Si-terminated centrosymmetric 31-layer slab of space group $P4/mmm$ (No. 123). Band structure calculations with $4f$ electrons treated as part of the frozen core are performed with the extended linear augmented plane wave (ELAPW) method [11] within the local density approximation (LDA) for the exchange-correlation functional using the full potential scheme of Ref. [12]. Based on our LDA calculations (we refer the reader to Ref. [13] for more details), we generated the relativistic $6 \times 6 \mathbf{k} \cdot \mathbf{p}$ Hamiltonian in the basis of \textit{ab initio} spinor wave functions $\Phi_{n\mu}$, which are unitary transformed to diagonalize the expectation value of the $z$-component of the total angular momentum $J = L + S$ ($n$ runs from 1 to 3, and $\mu = \uparrow$ or $\downarrow$ indicates the $z$-projection of $J$). The basis functions correspond to the surface states under study and are numbered in order of increasing energy at the $\overline{M}$ point, which is the reference $\mathbf{k}$-point of our second-order $\mathbf{k} \cdot \mathbf{p}$ expansion (see Supplementary Figure S6).

In the basis $\{\Phi_1\uparrow, \Phi_1\downarrow, \Phi_2\uparrow, \Phi_2\downarrow, \Phi_3\uparrow, \Phi_3\downarrow\}$, the Hamiltonian has the form:

$$H_{\mathbf{k}\mathbf{p}} = \begin{pmatrix}
\epsilon_1(k) & i\alpha_1 k_+ & i\delta k_+ & d(k) & Rk^2 & i\beta k_- \\
-i\alpha_1 k_+ & \epsilon_1(k) & -d^*(k) & i\delta k_- & -i\beta k_+ & Rk^2 \\
-i\delta k_- & -d(k) & \epsilon_2(k) & -i\alpha_2 k_+ & -i\delta k_+ & f(k) \\
d^*(k) & -i\delta k_+ & i\alpha_2 k_- & \epsilon_2(k) & f^*(k) & -i\delta k_+ \\
 Rk^2 & i\beta k_- & i\delta k_+ & f(k) & \epsilon_3(k) & i\alpha_3 k_- \\
-i\beta k_+ & Rk^2 & -f^*(k) & i\delta k_- & -i\alpha_3 k_+ & \epsilon_3(k)
\end{pmatrix}, \quad (1)$$

where $k_\pm = k_x \pm ik_y$, $k = \sqrt{k_x^2 + k_y^2}$, $\epsilon_i(k) = E_i + M_i k^2$, $d(k) = Dk_x^2 - Nk_y^2$, and $f(k) = Fk_x^2 - Gk_y^2$.

For the paramagnetic EuIr$_2$Si$_2$, the parameters in Eq. (1) are listed in Table I. By diagonalizing this Hamiltonian, $H_{\mathbf{k}\mathbf{p}} C^\lambda_{\mathbf{k}} = E^\lambda_{\mathbf{k}} C^\lambda_{\mathbf{k}}$, we obtained the model band dispersion curves $E^\lambda_{\mathbf{k}}$ and the six-dimensional vectors $C^\lambda_{\mathbf{k}}$ that yield the spinor solution $\tilde{\Phi}^\lambda_{\mathbf{k}} = \sum_{n\mu} C^\lambda_{n\mu\mathbf{k}} \Phi_{n\mu}$ – the state of the reduced Hilbert space of the $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian (1). The spin expectation value in this state is

$$\langle \mathbf{S}_{\mathbf{k}\lambda} \rangle = \frac{1}{2} \langle \tilde{\Phi}^\lambda_{\mathbf{k}} | \sigma | \tilde{\Phi}^\lambda_{\mathbf{k}} \rangle = \frac{1}{2} \sum_{n\mu\nu} C^\lambda_{n\mu\mathbf{k}} C^\lambda_{n\mu\mathbf{k}}^* \langle \mathbf{S} \rangle^n_{\mu\nu}. \quad (2)$$

Here, $[\mathbf{S}]^{n\mu}_{\nu} = \langle \Phi_{n\mu} | \sigma | \Phi_{n\nu} \rangle$ are the elements of the spin matrix $\mathbf{S} = (S_x, S_y, S_z)$ with the components (see Table I)

$$S_x = \begin{pmatrix}
\sigma_x & \bar{\sigma}_x & \sigma_{13} & \bar{\sigma}_{13} \\
\sigma_y & \bar{\sigma}_y & \sigma_{13} & \bar{\sigma}_{13} \\
\bar{\sigma}_x & \sigma_x & \bar{\sigma}_x & \sigma_{13} \\
\bar{\sigma}_y & \sigma_y & \bar{\sigma}_y & \sigma_{13}
\end{pmatrix}, S_y = \begin{pmatrix}
\sigma_y & \bar{\sigma}_y & \sigma_{13} & \bar{\sigma}_{13} \\
\sigma_x & \bar{\sigma}_x & \sigma_{13} & \bar{\sigma}_{13} \\
\bar{\sigma}_y & \sigma_y & \bar{\sigma}_y & \sigma_{13} \\
\bar{\sigma}_x & \sigma_x & \bar{\sigma}_x & \sigma_{13}
\end{pmatrix}, S_z = \begin{pmatrix}
\sigma_z & \bar{\sigma}_z & \sigma_{13} & \bar{\sigma}_{13} \\
\bar{\sigma}_z & \sigma_z & \bar{\sigma}_z & \sigma_{13} \\
\sigma_{13} & \bar{\sigma}_{13} & \sigma_z & \bar{\sigma}_z \\
\bar{\sigma}_{13} & \sigma_{13} & \bar{\sigma}_z & \sigma_z
\end{pmatrix}.$$
TABLE I. Parameters of the six-band $k \cdot p$ Hamiltonian (based on the LDA calculations for the 31-layer slab) for the Si-terminated surface of the paramagnetic EuIr$_2$Si$_2$. We use Rydberg atomic units: $\hbar = 2m_0 = e^2/2 = 1$.

<table>
<thead>
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<th>Parameter</th>
<th>Value 1</th>
<th>Value 2</th>
<th>Value 3</th>
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<tr>
<td>$E_1$</td>
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<td>2.67</td>
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<tr>
<td>$E_2$</td>
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<td></td>
<td>2.31</td>
</tr>
<tr>
<td>$E_3$</td>
<td>0.004</td>
<td></td>
<td>0.07</td>
</tr>
<tr>
<td>$\alpha_1$</td>
<td>0.223</td>
<td></td>
<td>1.63/1.63</td>
</tr>
<tr>
<td>$\alpha_2$</td>
<td>0.111</td>
<td></td>
<td>-0.30/-0.40</td>
</tr>
<tr>
<td>$\alpha_3$</td>
<td>-0.257</td>
<td></td>
<td>-0.85/0.76</td>
</tr>
<tr>
<td>$\delta$</td>
<td>-0.091</td>
<td></td>
<td>0.02/0.94</td>
</tr>
<tr>
<td>$\beta$</td>
<td>0.314</td>
<td></td>
<td>-0.31/0.64</td>
</tr>
<tr>
<td>$\vartheta$</td>
<td>-0.319</td>
<td></td>
<td>-0.07/0.33</td>
</tr>
<tr>
<td>$R$</td>
<td>-0.70</td>
<td></td>
<td>-0.31/0.64</td>
</tr>
</tbody>
</table>

where $\sigma_x$, $\sigma_y$, and $\sigma_z$ are the Pauli matrices and $\tilde{\sigma}_0 = i\sigma_0$, with $\sigma_0$ being the 2 × 2 identity matrix. The spin matrix also enters the term $H_{\text{EX}} = -J_{\text{ex}} \mathbf{M} \cdot \mathbf{S}$, which accounts for the exchange interaction of strength $J_{\text{ex}}$ with the magnetization $\mathbf{M}$ (see, e.g., Ref. [14]). Thus, to model the magnetic exchange interaction of surface-state electrons with the ferromagnetic rare-earth layer, we use this term and treat $\mathcal{J} = J_{\text{ex}} \mathbf{M}$ as a tunable parameter:

$$H_{\text{EX}} = - \begin{pmatrix} s_1^z \mathcal{J}_+ & s_1^z \mathcal{J}_- & s_2^z \mathcal{J}_+ & 0 & s_3^z \mathcal{J}_z & s_3^z \mathcal{J}_- \\ s_1^z \mathcal{J}_+ & -s_1^z \mathcal{J}_z & 0 & -s_2^z \mathcal{J}_- & s_3^z \mathcal{J}_z & s_3^z \mathcal{J}_- \\ s_2^z \mathcal{J}_- & 0 & s_2^z \mathcal{J}_z & s_2^z \mathcal{J}_+ & 0 & -s_3^z \mathcal{J}_+ \\ 0 & -s_2^z \mathcal{J}_+ & s_2^z \mathcal{J}_- & -s_2^z \mathcal{J}_z & 0 & -s_3^z \mathcal{J}_+ \\ s_3^z \mathcal{J}_z & s_3^z \mathcal{J}_- & 0 & s_3^z \mathcal{J}_z & s_3^z \mathcal{J}_- & -s_3^z \mathcal{J}_z \\ s_3^z \mathcal{J}_+ & -s_3^z \mathcal{J}_z & 0 & -s_3^z \mathcal{J}_+ & s_3^z \mathcal{J}_+ & -s_3^z \mathcal{J}_z \end{pmatrix},$$

where $\mathcal{J}_\pm = \mathcal{J}_x \pm i \mathcal{J}_y$. The resulting $k \cdot p$ Hamiltonian of our effective model, which we apply to study the modification of the spin-orbit split states by continuously varying the parameter $\mathcal{J}$, is the sum $H_{\text{Surf}} = H_{kp} + H_{\text{EX}}$. 
SUPPLEMENTARY REFERENCES

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