Supplementary Information
Non-Majorana modes in diluted spin chains proximitized to a superconductor

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Supplementary Note 1: Metallic vs superconducting STM tip

Figure S1: STS data taken with a normal metallic STM tip (grey) and a superconducting tip due to a Nb cluster at the apex obtained by tip indentations into the substrate. The left panel shows data on clean Nb. It shows that the gap size doubles when the tip is superconducting because of the convolution of tip and sample density of states, significantly enhancing the energy resolution. In the right panel depicting STS on a Cr adatom that features YSR resonance states inside the superconducting energy gap, the advantage of the enhanced energy resolution becomes clear since it enables a much more detailed analysis of the sharp in-gap peaks.
Supplementary Note 2: Cr adatom - Orbital character of YSR states

Figure S2: The figure shows spatial imaging of YSR resonances corresponding to different atomic orbitals. The topography data in the left panel has the same scale like the constant energy $dI/dU$ maps to the right, taken at energies 1.50 meV, 1.77 meV and 2.28 meV, from left to the right, indicated with dashed lines in the spectroscopy curve below taken on a single Cr adatom. Using STS, we observe the peak that corresponds to the Cr $d_{z^2}$ orbital as the most prominent one due to its longer extension into vacuum compared to the others.
Supplementary Note 3: 15 atom chain along $[1\bar{1}3]$

Figure S3: Spectroscopic mapping of the zero-energy mode appearing at boundary of a 15 adatom chain assembled along the $[1\bar{1}3]$ direction. The measurements have been acquired with a different microtip compared to the chain presented in the main text. The top panel report a full spectroscopy line across the chain, where dashed black lines correspond to the tip superconducting gap. The bottom panel highlights the rather symmetric electron- and hole- components of the zero-bias mode. To account for the asymmetric background at positive and negative energy (black lines in the top panel), the spectrum at the end of the chain has been normalized by subtracting the spectrum averaged over the bulk of the chain.
Supplementary Note 4: Surface preparation

**Figure S4:** Topographic images showing the areas around Cr chains. The first image correspond to the area where all chains presented in Fig. 2 in the main text have been assembled and measured. The second image correspond to the longest chain along the [1T3] direction. Because of the long range effect between Cr adatoms and the external environment, atomic manipulation techniques have been used to clean up the area before performing the experiments. The arrow indicate residual defects, all located some away from the chains, where their influence becomes negligible.
Supplementary Note 5: Spectroscopic data for short chains.

Figure S5: Scanning tunneling spectroscopy data for short chains. a-d Spectroscopic measurements across chains oriented along distinct crystallographic directions. e-h Spectra acquired by positioning the tip at the center (black line) and at the end (red line) of the chains. Dashed black lines correspond to the tip superconducting gap $\pm \Delta_{\text{tip}}$. i-l Topography (top panels) and spectroscopy map at zero energy (bottom panels) evidencing accumulation of spectra weight at the chains end.
Supplementary Note 6: Boundary modes in the metallic regime.

Although being in the diluted regime, where abrupt structural changes at the edge are small compared to densely packed chains, the existence of boundary modes localized at zero energy is confirmed by studying the same chains discussed in Fig. 2 once driving the system into the normal metallic regime, where Cr adatoms show a clear zero bias spectroscopic signature [1]. These results are illustrated in Figure S6. Its inspection reveals, also in the metallic regime, the existence of boundary modes which make the end of the chain distinct with respect to the center. This is evident by comparing the zero bias anomaly acquired by positioning the tip on each one of the adatom (see Fig.S6 d-f) and further highlighted in panels where spectra acquired in the center of the chains are directly overlapped to the spectra acquired on the last adatom (see Fig.S6g-i).

The localization of this boundary modes takes place on the same length scale as in the superconducting regime, i.e. the single atom level.

Figure S6: Distinct boundary behaviour in the metallic regime. a-c Topographic images five adatoms chains oriented along distinct crystallographic directions. d-f Spectroscopic mapping of the chain in the metallic regime, achieved by the application of a magnetic field of 1T perpendicular to the sample surface. A significant change in the zero bias anomaly is evident at the chain end, independently from their different crystallographic direction. These differences are highlighted in panels g-i.
Supplementary Note 7: Minimal tight-binding model

Supplementary Note 7.1: Tight-binding model

Here, we present the tight-binding model used in Sec. **Origin of zero energy boundary modes** of the main manuscript: the impurity chain, embedded in a two-dimensional superconductor, is described by the Hamiltonian

$$H_0 = \sum_{n,m} \left\{ \sum_{\nu,\nu'} \left\{ (4t-\mu)\delta_{\nu,\nu'} + J_{n,m}(\sigma_z)\nu,\nu' \right\} c_{n,m,\nu}^\dagger c_{n,m,\nu'} - \left\{ \sum_{\nu,\nu'} \left( t c_{n,m,\nu}^\dagger c_{n+1,m,\nu'}^\dagger + t c_{n,m,\nu}^\dagger c_{n,m+1,\nu'} \right) \delta_{\nu,\nu'} \right\} \right\} + \Delta c_{n,m,\nu}^\dagger c_{n,m,\nu}^\dagger + H.c. \right\}, \quad (S.1)$$

where $t$ and $\mu$ denote the matrix hopping element and the chemical potential, respectively. Furthermore, $\sigma_z$ is the Pauli $z$ matrix acting in spin space and $\delta_{\nu,\nu'}$ is the Kronecker delta. The operator $c_{n,m,\nu}^\dagger (c_{n,m,\nu}^\dagger)$ creates (annihilates) an electron with spin $\nu$ at the site $(n,m)$, where $n$ and $m$ denote the $x$ and $y$ position of the site in the two-dimensional lattice, respectively. The total number of lattice sites is given by $N_x \times N_y$. In addition, $J_{n,m}$ denotes the exchange coupling between the magnetic moments of the impurities and the spin of the itinerant electrons at the site $(n,m)$; and $\Delta$ is the superconducting gap. We set the lattice spacing $a$ to one. The system described by the Hamiltonian $H_0$ cannot enter a topological phase, except when Rashba spin-orbit interaction (SOI) is also present in the system. Such a Rashba SOI term is modelled by

$$H_R = \alpha \sum_{n,m} \left\{ \left( c_{n,m,\downarrow}^\dagger c_{n+1,m,\downarrow} - c_{n,m,\uparrow}^\dagger c_{n+1,m,\uparrow} + i c_{n,m,\downarrow} c_{n+1,m,\uparrow} - i c_{n,m,\uparrow} c_{n+1,m,\downarrow} \right) + H.c. \right\}, \quad (S.2)$$

where $\alpha$ denotes the Rashba SOI strength. We apply open boundary conditions and diagonalize the matrix $H = H_0 + H_R$, this yields the eigenvalues $E^l$ and the associated eigenvectors $\Psi^l$ consisting of the well known coherence factors $u^l_{n,m,\nu}$ and $v^l_{n,m,\nu}$. Furthermore, we define the local density of states (LDOS) at the site $(n,m)$ and at energy $\omega$ as

$$\rho(\omega, n, m) = \frac{1}{\pi} \text{Im} \left[ \sum_{l,\nu} \frac{|u^l_{n,m,\nu}|^2}{\omega - E_l + i\epsilon} + \frac{|v^l_{n,m,\nu}|^2}{\omega + E_l + i\epsilon} \right], \quad (S.3)$$

where the sum runs over the positive eigenvalues, and the parameter $\epsilon$ accounts for a broadening of the energy levels. We ignore states $\Psi^l$ with energies $E^l > \Delta$ for the calculation of $\rho$, since these states are energetically well separated from the lowest sub-gap states and we are mainly interested in the LDOS close to zero-energy.

Supplementary Note 7.2: Topological regime

In Sec. **Origin of zero energy boundary modes** of the main manuscript we focus on the trivial regime by neglecting SOI. Here, in the supplementary note we take SOI into account; the model can therefore enter the topological phase and could, in principle, host Majorana bound states (MBSs) under certain conditions. Fig. S7a shows the energies of a two-dimensional superconductor with a few adatoms, ordered as nearest neighbours, as a function of the $s-d$ exchange coupling strength $J$. The gap closing and reopening, which is predicted to accompany the topological phase transition, is only weakly pronounced due to the limited small number of adatoms. Furthermore, the restricted size of the system does not admit stable zero-energy states due to a strong overlap and hybridization of the MBSs. The results are slightly different in case of indirectly coupled adatoms, see Fig. S7b. The system exhibits a stable zero-energy state for some range of the exchange coupling. Moreover, the zero-energy state disappears at the exchange coupling $J = J_1$, where...
Supplementary Figure S7: Energies and probability densities of systems including SOI. (a, b, c) Energies $E$ as a function of the $s - d$ exchange coupling $J$. The black arrows above the panels indicate the critical exchange coupling $J_c$, for which a single bound state assumes zero energy. (d, e) Lowest energies as a function of the number of impurities $N$ at $J = J_c$. (f, g) Probability densities of the lowest state at $J = J_c$ for a system with $N = 10$ impurities. The parameters used in the plots are: $t = 1$, $\mu = 1$, $\alpha = 0.5$, $\Delta = 0.2$ and $J_c \approx 2.71$. Furthermore, we choose the lattice size $N_x \times N_y = 102 \times 60$, except in panel c in which we set $N_x \times N_y = 202 \times 50$. The chain is deposited at the $y$ coordinate $m_0 = 31$, except in panel e, where we set $m_0 = 26$. The impurities are separated by $L = a$ ($L = 2a$) in the panels a, d, and f (panels b, c, e, and g).

The gap closes and reopens. This closing and reopening, however, does not indicate a phase transition from trivial to topological. A second bulk gap closing and reopening, associated with this phase transition, appears only in much longer chains approximately at the exchange coupling for which the zero-energy pinning of the energetically lowest state starts, see Fig. S7c. In addition, in long chains it turns out that the gap closing and reopening at $J_1$ initializes a topological phase transition from topological to trivial rather than from trivial to topological phase.

In order to compare the numerical model with the experiment, in particular, with the data shown in Fig. 1b, we repeat the procedure described in the main manuscript: First, we consider a single magnetic impurity and calculate the energy as a function of the exchange coupling. The energy of the sub-gap state bound to the impurity is zero for $J = J_c$, indicated by the black arrow in Figs. S7a-c. Finally, we fix the exchange coupling to $J = J_c$ and calculate the energies as a function of the number of adatoms $N$, see Fig. S7d. The energies oscillate as a function of $N$ and the amplitude of these oscillations decays for values of $N$, which are much larger than the number of adatoms in the experiment, and a MBS stabilizes (one at
each end of the chain). Moreover, the MBS is energetically separated from the bulk states of the chain by a topological minigap $\Delta_T$. This minigap sets, among other parameters, the spatial localization of the MBS. A small minigap leads to spatially extended MBSs from each chain end which overlap and hybridize and form a non-zero energy fermionic bound state. Consequently, the formation of true MBSs in a system with a small minigap requires long chains. The experimental data, see Figs. 1b and 1d, reveal only a minor energetic separation between bulk and the edge states. It is therefore unlikely that the combination of a small minigap and a small number of adatoms support MBSs. Furthermore, the bulk modes of the chain are restricted in the experiment to a narrow energy interval, in theory, however, these states can assume energies $E_{\text{Bulk}}$ in the range $\Delta_T \leq E_{\text{Bulk}} < \Delta$, unless the Shiba bound states from the different adatoms are very weakly coupled. However, in the limit of almost uncoupled sub-gap states, the parameter space for the topological phase shrinks almost to zero. Finally, the oscillations of the lowest energy as a function of $N$ have not been observed in the experiment. Consequently, a topological origin of the zero-energy edge modes is unlikely. The experimental platform itself, however, represents an excellent starting point for the study of MBSs. Increasing the number of magnetic impurities might lead to further signatures, beside the zero-energy peak and its spectral weight on the ends of the chain, indicating the presence of MBSs. In addition, we numerically study the probability density of the lowest state in a short chain. The lowest state which transforms with growing chain length into MBSs has already higher weights on the ends of the chain, see Fig. S7f. For the sake of completeness, we also present the case of a chain with indirectly coupled adatoms, see Fig. S7e. The topological minigap is in this case much smaller, which agrees better with the energetic separation of end and bulk modes observed in the experiment. The energetic restriction to a narrow energy interval, however, still does not match with the numerical calculation. The probability density of a chain with ten impurities reveals again strong peaks at the ends of the chain, since the chain is effectively longer than the one build of nearest neighbour adatoms, see Fig. S7g. Finally, we conclude that only sufficiently long chains support the formation of topologically protected MBSs. This means, in particular, that the chain length needs to be much larger than the localization length of MBSs [2, 3].
Supplementary Note 8: Magnetic ground-state properties

To describe the magnetic structure of the Cr chains we use a generalized Heisenberg model,

\[ H = \sum_i e_i K_i e_i + \frac{1}{2} \sum_{ij} J_{ij} e_i \cdot e_j + \frac{1}{2} \sum_{ij} D_{ij} \cdot (e_i \times e_j) + \frac{1}{2} \sum_{ij} e_i J_{ij}^{aniso} e_j \]  

(S.4)

where \( i \) and \( j \) label the spins with a magnetic moment \( m_i = m_i e_i \), \( K_i \) is the magnetic on-site anisotropy, \( J_{ij} \) is the isotropic exchange interaction, \( D_{ij} \) is the Dzyaloshinskii-Moriya interaction, and \( J_{ij}^{aniso} \) is the symmetric anisotropic exchange. We assume that the on-site anisotropy is similar to that of the isolated Cr adatom,

\[ K = \begin{pmatrix} -0.02 & 0 & 0 \\ 0 & -0.17 & 0 \\ 0 & 0 & 0.19 \end{pmatrix} \text{meV} \]  

(S.5)

which was obtained from the method of constraining fields [4]. This assumption was tested by using the [001] chain as an example, for which we do not find any significant deviations from the isolated adatom values. The magnetic exchange interactions \( J_{ij} \) and \( D_{ij} \) of nearest neighbors and next-nearest neighbors are shown in Supplementary Figure S8a and b, respectively. As discussed in the main manuscript there are no significant edge effects observable, except for a small increase of the ferromagnetic coupling in case of the \([111]\) chain.

The magnetic structure is obtained by minimizing the Heisenberg model, which results in the angles shown in Figure 3 of the main manuscript and Supplementary Table S1. Due to the weakness of the DMI there is no significant non-collinearity present in the chains.

|         | \([001]\) | \([1\bar{1}3]\) | \([1\bar{1}1]\) | \([1\bar{1}0]\) |
|---------|-----------|-----------|-----------|-----------|
| \(\vartheta\) | \(\varphi\) | \(\vartheta\) | \(\varphi\) | \(\vartheta\) | \(\varphi\) |
| Atom 1  | 89.1°     | 90.0°     | 91.8°     | 260.5°    | 95.0°     | 268.8°    | 90.0°     | 270.0°    |
| Atom 2  | 90.5°     | 270.0°    | 89.9°     | 260.2°    | 92.2°     | 269.4°    | 90.0°     | 90.0°     |
| Atom 3  | 90.0°     | 90.0°     | 90.0°     | 260.2°    | 90.0°     | 269.4°    | 90.0°     | 270.0°    |
| Atom 4  | 89.6°     | 270.0°    | 90.1°     | 260.2°    | 87.8°     | 269.4°    | 90.0°     | 90.0°     |
| Atom 5  | 90.9°     | 90.0°     | 88.2°     | 260.5°    | 85.0°     | 268.8°    | 90.0°     | 270.0°    |

Supplementary Table S1: Magnetic ground states obtained from minimizing a Heisenberg model, eq. (S.4), with parameters shown in Supplementary Figure S8 and eq. (S.5).
Supplementary Figure S8: Magnetic exchange interactions of the investigated 5-atomic chains along the [001], [1\13], [1\11], and [1\10] directions. Shown are the nearest and next-nearest neighboring isotropic exchange interactions \(J\) (a) and Dzyaloshinskii-Moriya interactions \(D\) (b). The magnetic ground states obtained from these parameters are shown in Supplementary Table S1. The coordinate frame of the DMI vector is defined by the [1\10], [001], and [110] directions.
Supplementary Note 9: Multi-orbital tight-binding model and topological invariant

In the following, we use a tight-binding model with first-principles input to describe properties of the magnetic chains in the superconducting regime. We used the procedure in several works and refer the reader also to References [5, 6, 1], for additional details. Using the inverse of the Green function of the different chains obtained from first principles, we construct the hybridization function of the chain complexes
\[ H = E - G^{-1}(E), \]
which contains various information such as the hybridization strength with the surface, the crystal field splitting, the strength of spin-orbit coupling, and the strength of the direct hopping between the chain atoms. The Hamiltonian can be written as an effective tight-binding model, where the on-site part is given by (omitting the atom index \( i \) on the parameters)
\[ \mathcal{H}_i = \sum_{mm'} \sum_{s} (E_d \delta_{mm'} \delta_{ss'} + U \mathbf{e} \cdot \mathbf{\sigma}_{ss'} \delta_{mm'} + \lambda \mathbf{L}_{mm'} \cdot \mathbf{\sigma}_{ss'} + \Delta^{(re)}_{mm'} \delta_{ss'} + \Gamma \delta_{mm'} \delta_{ss'} + i \Delta^{(im)}_{mm'} \delta_{ss'} + \Delta^{(re)}_{mm'} \delta_{ss'}) \hat{c}_{im}^\dagger \hat{c}_{im'}, \]
and the inter-atomic hopping part is given by
\[ \mathcal{H}_{ij} = \sum_{mm'} \sum_{s} t_{mm'} c_{im}^\dagger c_{jm'} s. \]

\( E_d \) is the average energy of the \( d \)-orbitals with respect to the Fermi energy, \( 2U \) represents the exchange splitting of the magnetic moment pointing along \( e, \mathbf{\sigma} = (\sigma_x, \sigma_y, \sigma_z) \) is the vector of Pauli matrices, \( \lambda \) is the strength of the local spin-orbit coupling (-varies between 7 to 10 meV across the chains and directions), \( \mathbf{L} \) is the local orbital angular momentum operator, \( \Delta^{(re)} \) is an orbital dependent energy shift corresponding to the crystal field splitting, \( \Gamma \) and \( \Delta^{(im)} \) are non-hermitian contributions that result from the hybridization with the substrate, and \( t_{mm'} \) is the orbital-dependent hopping between atoms \( i \) and \( j \). The crystal field splitting \( \Delta^{(re)} \) of an isolated Cr adatom is shown in Supplementary Table S2, the nearest neighbor hoppings of all considered chains are shown in Supplementary Table S3, and more long-ranged hoppings in the [001] chain are shown in Supplementary Table S4.

We model the impact of the proximity-induced superconductivity on the magnetic chains by a \( s \)-wave pairing using the following form of the Bogouliubov-de-Gennes Hamiltonian,
\[ \mathcal{H}_{BdG} = \begin{pmatrix} \mathcal{H}_{\text{chain}} & \Delta_{mm'} \otimes i\sigma_y \\ -\Delta_{mm'} \otimes \sigma_y & -\mathcal{H}_{\text{chain}}^T \end{pmatrix}, \]

where \( \mathcal{H}_{\text{chain}} = \sum_i \mathcal{H}_i + \sum_{ij} \mathcal{H}_{ij} \).

From eqs. (S.7)-(S.9), we can directly define the Hamiltonian of an infinite one-dimensional chain in \( k \)-space using the Fourier transform,
\[ \mathcal{H}_{BdG}^{\text{BdG}}(k) = \sum_j \exp(ik(a_{ij} + R_{\mu\nu})) \mathcal{H}_{BdG}^{\text{BdG}}_{ij,\mu\nu}, \]

where the index \( i \) was left in for the sake of clarity and has no relevance for the infinite chain due to the translational invariance, \( \mu \) and \( \nu \) label the sites within a unit cell containing \( N_{uc} \) atoms, and \( a_{ij} \) and \( R_{\mu\nu} \) are the basis vectors. To simplify the later discussion we also assume that the lattice constant can be set to 1 resulting in \( a_{ij} = (j-i)N_{uc}, R_{\mu\nu} = \mu - \nu \) and \( k \in [-\pi/N_{uc}, \pi/N_{uc}] \).

Supplementary Note 9.1: YSR states of the isolated Cr adatom

In this section, we revisit the case of a single Cr adatom, which was extensively discussed in the Supplementary Notes of Ref. [5], to which we also refer for more details. Using the effective Hamiltonian, eq. (S.7),
gives direct access to the scattering phase shifts of each orbital $\delta_m^\pm$ and the related energies of the YSR states $\epsilon_m$,

$$\epsilon = \pm \cos \left( \delta_m^+ - \delta_m^- \right) \ .$$  \hspace{1cm} (S.11)

where $\Delta$ is the superconducting gap. In the case of the Cr adatom the YSR energies can be cast into the form

$$\frac{\epsilon_m}{\Delta} \approx \frac{1 - \alpha_m^2}{1 + \alpha_m^2} \quad \text{with} \quad \alpha_m = \frac{\Gamma_m U}{(E_m + U)(E_m - U)} \ .$$  \hspace{1cm} (S.12)

| $\Delta_{mm'}^{(re)}$ | $xy$ | $yz$ | $z^2$ | $xz$ | $x^2 - y^2$ |
|-----------------------|-----|-----|------|-----|----------|
| $xy$ | -0.07 | 0.00 | 0.00 | 0.00 | 0.00 |
| $yz$ | 0.00 | -0.08 | 0.00 | 0.00 | 0.00 |
| $z^2$ | 0.00 | 0.00 | 0.39 | 0.00 | -0.20 |
| $xz$ | 0.00 | 0.00 | 0.00 | -0.04 | 0.00 |
| $x^2 - y^2$ | 0.00 | 0.00 | -0.20 | 0.00 | -0.19 |

**Supplementary Table S2:** Crystal field splitting of the isolated Cr adatom in unit of [eV].

| [001] | $xy$ | $yz$ | $z^2$ | $xz$ | $x^2 - y^2$ |
|-------|-----|-----|------|-----|----------|
| $xy$ | -0.59 | 0.00 | 0.00 | 0.00 | 0.00 |
| $yz$ | 0.00 | -0.03 | 0.00 | 0.00 | 0.00 |
| $z^2$ | 0.00 | 0.00 | 0.10 | 0.00 | -0.01 |
| $xz$ | 0.00 | 0.00 | 0.00 | -0.02 | 0.00 |
| $x^2 - y^2$ | 0.00 | 0.00 | -0.01 | 0.00 | -0.16 |

| [113] | $xy$ | $yz$ | $z^2$ | $xz$ | $x^2 - y^2$ |
|-------|-----|-----|------|-----|----------|
| $xy$ | -0.10 | 0.00 | 0.17 | 0.00 | 0.09 |
| $yz$ | 0.00 | -0.13 | 0.00 | 0.08 | 0.00 |
| $z^2$ | 0.17 | 0.00 | 0.15 | 0.00 | 0.01 |
| $xz$ | 0.00 | 0.08 | 0.00 | -0.25 | 0.00 |
| $x^2 - y^2$ | 0.09 | 0.00 | 0.01 | 0.00 | 0.05 |

| [111] | $xy$ | $yz$ | $z^2$ | $xz$ | $x^2 - y^2$ |
|-------|-----|-----|------|-----|----------|
| $xy$ | 0.32 | -0.00 | 0.02 | 0.00 | -0.02 |
| $yz$ | 0.00 | 0.03 | 0.00 | -0.03 | 0.00 |
| $z^2$ | 0.02 | 0.00 | 0.08 | -0.01 | 0.10 |
| $xz$ | 0.00 | -0.03 | -0.01 | 0.14 | 0.00 |
| $x^2 - y^2$ | -0.02 | 0.00 | 0.10 | 0.00 | 0.07 |

| [110] | $xy$ | $yz$ | $z^2$ | $xz$ | $x^2 - y^2$ |
|-------|-----|-----|------|-----|----------|
| $xy$ | -0.25 | 0.00 | 0.00 | 0.0 | 0.00 |
| $yz$ | 0.00 | -0.03 | 0.00 | 0.0 | 0.00 |
| $z^2$ | 0.00 | 0.00 | 0.15 | 0.0 | -0.01 |
| $xz$ | 0.00 | 0.00 | 0.00 | 0.1 | 0.00 |
| $x^2 - y^2$ | 0.00 | 0.00 | -0.01 | 0.0 | -0.05 |

**Supplementary Table S3:** Hopping matrices for the chains along [001], [113], [111], and [110] directions. Shown are the nearest-neighbor Cr-Cr hopping matrices in unit of [eV].
Supplementary Table S4: Hopping matrices for the [001] chain and the second, the third and the fourth neighbor hopping in units of [eV].

|   | $t_{13}$ | xy | yz | $z^2$ | $x^2 - y^2$ | $x^2 - y^2$ |
|---|---------|----|----|------|--------------|--------------|
| xy | 0.22    | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| yz | 0.00    | 0.03 | 0.00 | 0.00 | 0.00 | 0.00 |
| $z^2$ | 0.00 | 0.00 | 0.14 | -0.00 | 0.02 |
| $x^2 - y^2$ | 0.00 | 0.00 | -0.00 | 0.04 | 0.00 |

|   | $t_{14}$ | xy | yz | $z^2$ | $x^2 - y^2$ | $x^2 - y^2$ |
|---|---------|----|----|------|--------------|--------------|
| xy | -0.16   | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| yz | 0.00    | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| $z^2$ | 0.00 | 0.00 | -0.12 | 0.00 | -0.02 |
| $x^2 - y^2$ | 0.00 | 0.00 | 0.00 | -0.04 | 0.00 |

|   | $t_{15}$ | xy | yz | $z^2$ | $x^2 - y^2$ | $x^2 - y^2$ |
|---|---------|----|----|------|--------------|--------------|
| xy | 0.15    | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 |
| yz | 0.00    | -0.04 | 0.00 | 0.00 | 0.00 | 0.00 |
| $z^2$ | 0.00 | 0.00 | 0.07 | 0.00 | 0.01 |
| $x^2 - y^2$ | 0.00 | 0.00 | 0.00 | 0.02 | 0.00 |

Supplementary Figure S9: Energy of the YSR states $E_{YSR}/\Delta$ of the isolated Cr adatom as function of the energy of the orbital $E_d$ and its hybridization strength $\Gamma$. Shown are the full parametrizations obtained from first principles (triangles), as well as the energies obtained from a reduced hybridization $\tilde{\Gamma} = 0.8\Gamma$ (circles).

The YSR energies obtained from this scheme using the first-principles parameters are shown as triangles in Supplementary Figure S9. To match the experimentally observed energy spectrum of the YSR states of Ref. [5], we renormalize the hybridization $\Gamma$ by a factor of 0.8, which is shown by the circles in Supplementary Figure S9. With this approach, we ensure that we obtain the $z^2$-like YSR state near zero energy, and we also obtain the correct order of the experimentally observed YSR states. The need for this renormalization can have several origins, e.g. an overestimation of the geometric relaxation of the adatom towards the surface could increase the hybridization or the general parameterization could be renormalized in the superconducting regime, which we do not address from first principles.
Supplementary Note 9.2: Magnetism and topological invariant

For the classification of the topology of the used tight-binding model, special care has to be taken with respect to the non-hermicity. A detailed discussion of the topology in non-hermitian physics can be found, for example, in Ref. [7]. The particle-hole symmetry of the non-hermitian Hamiltonian defined in eq. (S.7) is reflected as

\[ C^{-1}H^T(k)C_- = -H(-k) \quad \text{with} \quad C_-C^*_+ = +1 \quad . \]  

(S.13)

\( C_- \) is given by \( \tau_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \) acting in the particle-hole space. For this case, a \( \mathbb{Z}_2 \) invariant \( \nu \in \{0,1\} \) can be defined by

\[ (-1)^\nu = \frac{\text{Pf}[H(\pi)C_-]}{\text{Pf}[H(0)C_-]} \exp \left[ -\frac{1}{2} \int_0^\pi dk \log \det[H(k)C_-] \right] \quad , \]  

(S.14)

where Pf denotes the Pfaffian. This definition for the topological invariant is used in the phase diagrams shown in Figure 5b of the main manuscript.
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