Supplementary material for: A universal route to chiral Ising superconductivity in monolayer TaS_2 and $NbSe_2$

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(Dated: November 25, 2025)

CONTENTS

I. Parametrization of the LCAO tight-binding model 1
II. Matrix elements of the screened interaction 1
III. Experimental setup and methods 1
IV. Comparison to experimental STS data 2
References 2

I. PARAMETRIZATION OF THE LCAO TIGHT-BINDING MODEL

We present details regarding the multi-orbital tight-binding calculation of the relevant low-energy bands of 1H-$NbSe_2$ and 1H-TaS_2 shown in Fig. 2 of main text. For the former, the bands are obtained from a linear combination of the three atomic d-orbitals of Nb $4d_{z^2} = 4d_{2,0}$ and $4d_{2,\pm 2} = 4d_{x^2-y^2} \pm i4d_{xy}$. In TaS_2 the orbitals come from the next shell, and hence we consider $5d_{z^2} = 5d_{2,0}$ and $5d_{2,\pm 2} = 5d_{x^2-y^2} \pm i5d_{xy}$. For the functional form of the orbitals we use the one of the hydrogen atom with effective nuclear charge Z_{eff} from [1]. The tight-binding parametrization by He *et al.* [2] is adopted for 1H-$NbSe_2$, but similar results are obtained from the one reported by Kim and Son [3]. For 1H-TaS_2 we rely on the results by [4].

II. MATRIX ELEMENTS OF THE SCREENED INTERACTION

In the Bloch basis the matrix elements of the screened interaction have the form

$$\langle \mathcal{Q}(\boldsymbol{k}+\boldsymbol{q}), \sigma; \mathcal{Q}(\boldsymbol{k}'-\boldsymbol{q}), \sigma' | \hat{V} | \boldsymbol{k}, \sigma; \boldsymbol{k}', \sigma' \rangle = \frac{1}{N\Omega} \sum_{\boldsymbol{G},\boldsymbol{G}'} V^{\text{2D,RPA}}_{\boldsymbol{G},\boldsymbol{G}'}(\boldsymbol{q}; 0^+) \mathcal{F}^{\sigma}_{\boldsymbol{k}+\boldsymbol{q},\boldsymbol{k}}(-\boldsymbol{G}) \mathcal{F}^{\sigma'}_{\boldsymbol{k}'-\boldsymbol{q},\boldsymbol{k}'}(\boldsymbol{G}')\,, \tag{B1}$$

where $\boldsymbol{k}, \boldsymbol{k}'$ and $\boldsymbol{q}$ are in-plane momenta restricted to the first Brillouin zone. Furthermore, the projector $\mathcal{Q}$ ensures that the scattered momenta are folded back into the first Brillouin zone as well. We omit to indicate the projector $\mathcal{Q}$ in the following for simplicity. The Bloch overlaps $\mathcal{F}$ are defined as

$$\mathcal{F}^{\sigma}_{\boldsymbol{k},\boldsymbol{k}'}(\boldsymbol{G}) = \int_{\mathcal{V}_p} d\boldsymbol{r} e^{-i\boldsymbol{G}\cdot\boldsymbol{r}} \boldsymbol{u}^{\dagger}_{\sigma,\boldsymbol{k}}(\boldsymbol{r}) \boldsymbol{u}_{\sigma,\boldsymbol{k}'}(\boldsymbol{r})\,, \tag{B2}$$

with $\boldsymbol{u}_{\sigma,\boldsymbol{k}}$ Bloch spinors, and the unit cell volume $\mathcal{V}_p$.

The matrix elements in Eq. (B1) are in general complex, with a strongly fluctuating phase resulting from the product of the Bloch overlaps. This tends to favor pairing between time-reversed states, whose Bloch spinors obey the relation $\boldsymbol{u}_{\bar{\sigma},\bar{\boldsymbol{k}}} = \boldsymbol{u}^*_{\sigma,\boldsymbol{k}}$, with $-\boldsymbol{k} =: \bar{\boldsymbol{k}}, -\sigma =: \bar{\sigma}$, resulting in a summation of real and positive contributions. We hence retain only the scattering between time-reversal partners $(\boldsymbol{k}, \sigma; \bar{\boldsymbol{k}}, \bar{\sigma})$, and consider the following interaction matrix elements $V_{\boldsymbol{k},\boldsymbol{k}',\sigma} := \langle \boldsymbol{k}', \sigma'; \bar{\boldsymbol{k}}', \bar{\sigma}' | \hat{V} | \boldsymbol{k}, \sigma; \bar{\boldsymbol{k}}, \bar{\sigma} \rangle$.

III. EXPERIMENTAL SETUP AND METHODS

TaS_2 monolayers$_2$ were grown on highly oriented pyrolytic graphite (HOPG) using molecular beam epitaxy (MBE). After growth, the sample was inserted into the low-temperature STM (Unisoku USM-1300) housed in the same ultra-high vacuum system and subsequent experiments were performed at $T = 350$ mK. The dI/dV spectra were recorded by standard lock-in detection while sweeping the sample bias in an open feedback loop configuration, with a peak-to-peak bias modulation specified for each measurement and at a frequency of 911 Hz. The dI/dV spectra were recorded on extended monolayers to eliminate possible island size dependence from the measurements. The data shown in main text Fig. 6 are normalized by the averaged dI/dV.

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IV. COMPARISON TO EXPERIMENTAL STS DATA

For tunneling spectroscopy with a normal metal tip, the differential conductance is well approximated by

$$G(V) \approx \sum_{\pi} C_\pi G_\pi(V)\,, \tag{D1}$$

$$G_\pi(V) = \int_{-\infty}^{\infty} \mathrm{d}E\, D_\pi(E) \left(-\frac{\partial f(E+eV)}{\partial E}\right)\,, \tag{D2}$$

where the couplings C_π, account for the spectral function of the tip and the tunneling overlap between the tip's evanescent states and quasiparticle states from the superconductor on a given Fermi surface $\pi = K, \Gamma$. The tunneling to each of the Fermi surfaces represents a distinct transport channel, whose strength is given by the sum over the local quasiparticle density of states

$$D_\pi(E) = \sum_{\boldsymbol{k}\in\pi,\sigma} D_{\boldsymbol{k},\sigma}(E)\,. \tag{D3}$$

The latter are modeled by assuming a BCS form factor modifying the local density of states $\rho_{\boldsymbol{k},\sigma}$ in the normal conducting state. In order to reproduce the experimentally measured critical temperature of $T_c = 1$ K [5], we used a gap obtained by solving the gap equation with the interaction potential $\hat{V}$ multiplied by a factor γ, which accounts, for example, for charge transfer from the substrate or an additional phonon-mediated interaction [6, 7], which have not been explicitly included in our calculation.

Due to the different orbital composition of the quasiparticle states at the K, K' valleys and at the Γ surface [8], the coupling constant will be in general different. In particular, since the $d_{2,0}$ orbital extends farthest out of plane and the STM favors states with small in-plane momentum [9, 10], we expect $C_\Gamma \gg C_K$. For low temperature, the form of the gap on each separate Fermi surface results in characteristic signatures in the differential conductance of the respective transport channel. We use as a fit function this superposition of the differential conductance of the individual transport channels. For the gaps on the Fermi surfaces $\Delta_{\boldsymbol{k},\sigma}$, we use the form of the gaps as found from main text Eq. (3) at the experimental temperature T_{exp} and allow for a fit of the amplitude by a common rescaling of all $\Delta_{\boldsymbol{k},\sigma}$ by a parameter A as $\Delta_{\boldsymbol{k},\sigma} \to A\Delta_{\boldsymbol{k},\sigma}$. The latter is introduced to take care of the uncertainty of the actual experimentally realized T_{c} and the rescaling factor γ of the interaction.

To qualitatively account for additional sources of broadening in the experiment, we fit with an effective temperature T_{eff} in the calculation of the derivative in Eq. (D2), about twice the recorded base temperature of the STM. To account for offsets in the calibration, we further allow for both a small constant offset G_0 in the measured conductivity and $V_0 = 0.009$ mV in the recorded voltage. For the fitting we use the trust region reflective algorithm as implemented in SciPy's "curve_fit" routine. We consider a range of ± 0.5 mV containing the main coherence peaks. The resulting fit parameters are listed in Table I. The best fits for all the solutions are obtained by considering strongly selective coupling of the tip to the Γ pockets.

Gap	C_K , C_Γ (eV Å^2)	A	G_0	T_{eff}/T	γ
chiral	1.607, 15.041	1.110	0.173	2.175	9.298
nematic	0.000, 17.235	1.156	0.174	1.815	9.298
$s+f$	1.890, 18.333	1.017	0.000	1.804	8.777

Table I. Coefficients of best fit between the theoretical prediction of the differential conductance in the possible superconducting phases and the experiment as shown in Fig. 6 (b), (c). The best fit for each solution is achieved by selective coupling to the Γ pocket.

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