Supplemental Material
Observation of short-range Yu-Shiba-Rusinov states with threefold symmetry in layered superconductor 2H-$NbSe_2$

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I. GENERAL FORMULATION OF YSR STATE

Consider a superconductor described by the Hamiltonian $\hat{H}$ and the impurity potential $\hat{V}_{imp}$ which has a finite real space support $\mathcal{S}$, namely $V_{imp}(\mathbf{r}) \neq 0$ for $\mathbf{r} \in \mathcal{S}$. The YSR state $|\psi\rangle$ satisfies the Schrödinger equation

$$\hat{G}^{-1}(E)\,|\psi\rangle = \hat{V}_{imp}\,|\psi\rangle\,, \tag{1}$$

with $\hat{G}^{-1}(E) = E - \hat{H}$ denote the inverse Green function of the superconductor. Thus, we have

$$|\psi\rangle = \hat{G}(E)\hat{V}_{imp}\,|\psi\rangle\,. \tag{2}$$

Thus, we have for $\mathbf{r}, \mathbf{r}' \in \mathcal{S}$, the wave function of the YSR state satisfies

$$\psi(\mathbf{r}) = \sum_{\mathbf{r}'} G(E, \mathbf{r} - \mathbf{r}')V_{imp}(\mathbf{r}')\psi(\mathbf{r}'), \tag{3}$$

and the YSR state energy E_s is determined from the condition $\det(\mathbf{F}(E = E_s)) = 0$, where the matrix elements of the matrix $\mathbf{F}(E)$ are

$$F(E, \mathbf{r}, \mathbf{r}') = \delta_{\mathbf{r},\mathbf{r}'}\mathbb{I} - G(E, \mathbf{r} - \mathbf{r}')V_{imp}(\mathbf{r}'), \tag{4}$$

where $G(E, \mathbf{r} - \mathbf{r}')$ is the real space representation of $\hat{G}(E)$ and $\mathbb{I}$ is the identity matrix of the same size as $G(E, \mathbf{r} - \mathbf{r}')$.

II. YSR STATE IN A 3D LAYERED SUPERCONDUCTOR

Let us consider a system consisting of layers of 2D BCS superconductors. Let us further assume the 2D layers can be described by a continuum Bogoliubov-de Gennes Hamiltonian, and there exists an interlayer coupling (e.g. due to van der Waals interaction) that couples nearest neighbor layers. The second quantized Hamiltonian for this system can be written as

$$\mathcal{H} = \sum_j \int \frac{dk^2}{(2\pi)^2} \psi_j^\dagger(\mathbf{k}) H_{layer}(\mathbf{k})\psi_j(\mathbf{k}) + (t\psi_j^\dagger(\mathbf{k})\tau_z\psi_{j+1}(\mathbf{k}) + h.c.), \tag{5}$$

$$H_{layer}(\mathbf{k}) = (\frac{\mathbf{k}^2}{2m} - \mu)\tau_z + \Delta\tau_x \equiv \xi_k\tau_z + \Delta\tau_x, \tag{6}$$

where $\mathbf{k}$ is the 2D momentum within one layer, μ is the chemical potential, Δ is the superconducting pairing potential, j is the layer index, $\tau_{x,z}$ are Pauli matrices in the Nambu space, t is the coupling between neighboring layers, $\psi_j^\dagger$ and ψ_j are the corresponding Nambu spinors consisting of the electron creation and annihilation operators at momentum $\mathbf{k}$.

Let us assume the magnetic impurity only couples to a single layer of the superconductor, and compute the wave function amplitude of the YSR state within this layer. Using the general formulation introduced in the previous section, we first compute the Green function $G(E, \mathbf{r}, n)$, which describes the amplitude when a quasiparticle of energy E propagates for a 2D distance parallel to the layers and crosses n layers. Thus, the wave function amplitude at position $\mathbf{r}$ of the mth layer$\psi(\mathbf{r}, m)$ then satisfies

$$\psi(\mathbf{r}, m) = \sum_{\mathbf{r}'} G(E, \mathbf{r} - \mathbf{r}', m)V_{imp}(\mathbf{r}')\psi(\mathbf{r}', 0), \tag{7}$$

where we set the impurity potential $V_{imp}(\mathbf{r})$ only exist in the 0th layer. Particularly, we are interested in the behavior of the wave function within the layer that contains the magnetic impurity, namely $\psi(\mathbf{r},0)$. For a point impurity at $\mathbf{r}=0$, we have

$$\psi(\mathbf{r},0) = G(E,\mathbf{r},0)V_{imp}(0)\psi(0,0). \tag{8}$$

Thus, the spatial profile of $\psi(\mathbf{r},0)$ is closely related to the spatial profile of $G(E,\mathbf{r},0)$, which we will compute in the following.

The Green function $G(E,\boldsymbol{r},n)$ can be obtained from its momentum space representation

$$\begin{aligned} G(E,\mathbf{r},n) &= \frac{1}{(2\pi)^3}\int_{-\pi}^{\pi} dk_z e^{ik_z n}\int d\mathbf{k}^2 e^{i\mathbf{k}\cdot\mathbf{r}}\frac{E+(\xi_k+2t\cos k_z)\tau_z+\Delta\tau_x}{E^2-(\xi_k+2t\cos k_z)^2-\Delta^2} \\ &\simeq \frac{1}{2\pi}\int_{-\pi}^{\pi} dk_z e^{ik_z n}\frac{\nu_{2d}}{2\pi}\int_{-\infty}^{\infty}d\xi\int_0^{2\pi}d\theta e^{ikr\cos\theta}\frac{E+\xi\tau_z+\Delta\tau_x}{E^2-\xi^2-\Delta^2} \\ &= \frac{\nu_{2d}}{2}\int_{-\pi}^{\pi}dk_z e^{ik_z n}\left[-\Re[f(r,k_z)]\frac{(E+\Delta\tau_x)}{\sqrt{\Delta^2-E^2}}+\Im[f(r,k_z)]\tau_z\right]. \end{aligned} \tag{9}$$

Here, ν_{2d} the 2D density of states at the Fermi energy, $r=|\mathbf{r}|$, k_z is the Bloch momentum perpendicular to the layers when periodic boundary condition is assumed, and

$$f(r,k_z) = J_0(k_F(1-\frac{2t}{\mu}\cos k_z)r+ik_s r)+iH_0(k_F(1-\frac{2t}{\mu}\cos k_z)r+ik_s r),$$

where $k_F=\sqrt{2m\mu}$ is the Fermi momentum, $k_s=k_F\sqrt{\Delta^2-E^2}/\mu$. J_0 and H_0 are Bessel function and Hankel function, respectively.

The real space single-particle Green function, which describes the amplitude when a quasiparticle of energy E propagates for a 2D distance parallel to the layers and crosses n layers, can be written as

In the asymptotic limit with $k_F r\gg 1$, we have

$$f(r,k_z)\simeq\sqrt{\frac{2}{\pi k_F r(1-\frac{2t}{\mu}\cos k_z)}}\exp\left[ik_F r(1-\frac{2t}{\mu}\cos k_z)-\frac{i\pi}{4}-k_s r\right]+\frac{2i}{\pi k_F r(1-\frac{2t}{\mu}\cos k_z)}. \tag{10}$$

We have

$$G(E,\mathbf{r},0)=\nu_{2d}\left[\frac{(E+\Delta\tau_x)}{\sqrt{\Delta^2-E^2}}f_0(r)+f_1(r)\tau_z\right] \tag{11}$$

with

$$\begin{aligned} f_0(r) &= -\frac{1}{2}\sqrt{\frac{2}{\pi k_F r}}e^{-k_s r}\int_{-\pi}^{\pi}dk_z\sqrt{\frac{1}{1-\frac{2t}{\mu}\cos k_z}}\cos(k_F r(1-\frac{2t}{\mu}\cos k_z)-\frac{\pi}{4}) \\ &\simeq -\frac{1}{2}\sqrt{\frac{2}{\pi k_F r}}e^{-k_s r}\int_{-\pi}^{\pi}dk_z(1+\frac{t}{\mu}\cos k_z)\cos(k_F r(1-\frac{2t}{\mu}\cos k_z)-\frac{\pi}{4}) \\ &= -\sqrt{\frac{2\pi}{k_F r}}e^{-k_s r}\left[J_0(2k_F r\frac{t}{\mu})\cos(k_F r-\frac{\pi}{4})+\frac{t}{\mu}J_1(2k_F r\frac{t}{\mu})\sin(k_F r-\frac{\pi}{4})\right] \end{aligned} \tag{12}$$

$$\begin{aligned} f_1(r) &= \frac{1}{2}\sqrt{\frac{2}{\pi k_F r}}e^{-k_s r}\int_{-\pi}^{\pi}dk_z\sqrt{\frac{1}{1-\frac{2t}{\mu}\cos k_z}}\sin(k_F r(1-\frac{2t}{\mu}\cos k_z)-\frac{\pi}{4})+\frac{1}{\pi k_F r}\int_{-\pi}^{\pi}dk_z\frac{1}{(1-\frac{2t}{\mu}\cos k_z)} \\ &\simeq \frac{1}{2}\sqrt{\frac{2}{\pi k_F r}}e^{-k_s r}\int_{-\pi}^{\pi}dk_z(1+\frac{t}{\mu}\cos k_z)\sin(k_F r(1-\frac{2t}{\mu}\cos k_z)-\frac{\pi}{4})+\frac{2}{k_F r\sqrt{1-4t^2/\mu^2}} \\ &= \sqrt{\frac{2\pi}{k_F r}}e^{-k_s r}\left[J_0(2k_F r\frac{t}{\mu})\sin(k_F r-\frac{\pi}{4})-\frac{t}{\mu}J_1(2k_F r\frac{t}{\mu})\cos(k_F r-\frac{\pi}{4})\right]+\frac{2}{k_F r\sqrt{1-4t^2/\mu^2}}. \end{aligned} \tag{13}$$

At small distance with $2k_F rt/\mu\ll 1$, we have

$$f_0(r)\simeq -\left[\sqrt{\frac{2\pi}{k_F r}}\cos(k_F r-\frac{\pi}{4})+(\frac{t}{\mu})^2\sqrt{2\pi k_F r}\sin(k_F r-\frac{\pi}{4})\right]e^{-k_s r} \tag{14}$$

$$f_1(r)\simeq\left[\sqrt{\frac{2\pi}{k_F r}}\sin(k_F r-\frac{\pi}{4})-(\frac{t}{\mu})^2\sqrt{2\pi k_F r}\cos(k_F r-\frac{\pi}{4})\right]e^{-k_s r}+\frac{2}{k_F r\sqrt{1-4t^2/\mu^2}}, \tag{15}$$

Table I. Tight-binding model parameters (in meV)

ξ_0	t_1	t_2	t_3	t_4	t_5
203.0	46.0	257.5	4.4	−15.0	6.0

which implies the wave function $\psi(\mathbf{r},0)$ decays as $\propto 1/\sqrt{r}$, similar to the case of a 2D system.

At a larger distance when $2k_F rt/\mu \gg 1$, we have

$$f_0(r) \simeq -\frac{1}{k_F r}\sqrt{\frac{2\mu}{t}}e^{-k_s r}\left[\cos(2k_F r\frac{t}{\mu}-\frac{\pi}{4})\cos(k_F r-\frac{\pi}{4})+\frac{t}{\mu}\cos(2k_F r\frac{t}{\mu}-\frac{3\pi}{4})\sin(k_F r-\frac{\pi}{4})\right] \tag{16}$$

$$f_1(r) \simeq \frac{1}{k_F r}\sqrt{\frac{2\mu}{t}}e^{-k_s r}\left[\cos(2k_F r\frac{t}{\mu}-\frac{\pi}{4})\sin(k_F r-\frac{\pi}{4})-\frac{t}{\mu}\cos(2k_F r\frac{t}{\mu}-\frac{3\pi}{4})\cos(k_F r-\frac{\pi}{4})\right]. \tag{17}$$

In this case, we have the wave function $\psi(\mathbf{r},0)$ decays as $\propto 1/r$, similar to the case of a 3D system.

The crossover between the two types of decay behavior happens at a distance $r_c \sim v_F/(2t)$, with Fermi velocity $v_F = \mu/k_F$. Thus, when the interlayer coupling is strong, we are able to have a small r_c, which cuts off the long-range decaying wave function.

III. THREE-FOLD SYMMETRY FROM A TIGHT-BINDING MODEL

In this section, we describe the detailed tight-binding simulation of a YSR state with a threefold symmetry. We take the 1-band model on a triangular lattice with up to fifth nearest neighbor hopping, which faithfully captures the lowest-lying Nb 4d-derived states of 2H-NbSe2 [1], For convenience, we list the detail tight-binding parameters below. The band dispersion has the following form

$$\begin{aligned}\xi(\mathbf{k}) = \xi_0 &+ t_1(2\cos\xi\cos\eta+\cos 2\xi)+t_2(2\cos 3\xi\cos\eta+\cos 2\eta)+t_3(2\cos 2\xi\cos 2\eta+\cos 4\xi)\\ &+t_4(\cos\xi\cos 3\eta+\cos 5\xi\cos\eta+\cos 4\xi\cos 2\eta)+t_5(2\cos 3\eta\cos 3\xi+\cos 6\xi)\end{aligned} \tag{18}$$

with $\xi = \frac{1}{2}k_x a$ and $\eta = \frac{\sqrt{3}}{2}k_y a$, and a is the triangular lattice spacing. The parameters in this model are listed in Table I.

In the tight-binding model, the electrons hop on the lattice with coordinates $\mathbf{r}_{nm} = n\mathbf{a}_1 + m\mathbf{a_2}$, where $\mathbf{a}_1 = (1,0)a$ and $\mathbf{a}_2 = (-1,\frac{\sqrt{3}}{2})a$ are the two lattice vectors. The Bogoliubov–de Gennes Hamiltonian for the corresponding superconductor can be written as

$$H_{1L}(\boldsymbol{k}) = \xi(\boldsymbol{k})\tau_z + \Delta\tau_x, \tag{19}$$

and we take$\Delta = 1$meV in this work.

The impurity potential can be written as

$$V_{imp}(\mathbf{r}) = (-JS + K\tau_z)f(\mathbf{r}), \tag{20}$$

where the first and second terms correspond to magnetic and potential scatterings, respectively. The scalar function $f(\mathbf{r})$ determines the range of the interaction. In Ref. [1], $f(\mathbf{r}) = \delta_{\mathbf{r},\mathbf{r}_{00}}$, namely, the impurity potential is only on one of the triangular site. This produces a YSR state with six-fold degeneracy, see Fig. 3(a) of the main text.

To produce a three-fold symmetric YSR state, we choose $f(\mathbf{r}) = \delta_{\mathbf{r},\mathbf{r}_{00}} + \delta_{\mathbf{r},\mathbf{r}_{-1,0}} + \delta_{\mathbf{r},\mathbf{r}_{01}}$, which essentially introduces an impurity potential on the three nearest sites, simulating a hollow site adatom, as shown in Fig.3 (b) of the main text. Using the general formalism described in Sec. I, one can compute the energy of the YSR state, as well as the corresponding wave function. To produce Fig. 3(a) and (b) in the main text, we set $JS = K = 200$meV.

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