Supporting Information for Cobalt-Dimer Nitrides: A Potential Novel Family of High Temperature Superconductors

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COMPUTATIONAL METHODS

We employ the Vienna *ab initio* simulation package (VASP) code[1] with the projector augmented wave (PAW) method[2] to perform DFT calculations. The Perdew-Burke-Ernzerhof (PBE)[3] exchange-correlation functional is used in our calculations. The kinetic energy cutoff is set to be 600 eV for expanding the wave functions into a plane-wave basis in VASP calculations. We use the primitive cell of $BaCo_2N_2$ to calculate its electronic structure. The crystal structure is fully relaxed while the force convergence criterion is 0.005 eV/Å. The energy convergence criterion is 10^{-7} eV and the Γ-centered **k**-mesh is $12 \times 12 \times 12$. The Co_2N_2 monolayer is extracted from the relaxed bulk $BaCo_2N_2$ with 40Å thick vacuum layer and its Γ-centered **k**-mesh is $16 \times 16 \times 1$.

We employ Wannier90[4, 5] to calculate maximally localized Wannier functions in $BaCo_2N_2$, which perfectly reproduces DFT-calculated band structure around the Fermi level. The initial projectors are Co's d-orbitals with local symmetric $X - Y$ coordinate.

In the study of the magnetism of $BaCo_2N_2$, the DFT+U method in the formulation of Liechtenstein et al. [8] is employed to describe the associated electron-electron correlation effect and J is fixed as $\frac{U}{4}$. To estimate the magnetic exchange between two NN dimers, the energies of different magnetic states are calculated with the relaxed structure of paramagnetic $BaCo_2N_2$.

EXPLICIT FORM OF 4-BAND TB MODEL

The 4-band effective tight-binding (TB) model before unfolding with the basis of the d_{XY} and d_{YZ} orbitals is given by

$$H_t^{4-band} = \sum_{k\alpha\beta ij} \varepsilon_k^{\alpha\beta ij} d_{k\alpha i}^\dagger d_{k\beta j} + \sum_{ki} e_\alpha d_{k\alpha i}^\dagger d_{k\alpha i}, \tag{1}$$

where α/β is the orbital index, i/j is the site index and e_α denotes the crystal field energy of each orbital. The corresponding expressions for the hopping energies in Eq. (1) are given by

$$\varepsilon_k^{\alpha\alpha AA} = \varepsilon_k^{\alpha\alpha BB} = 2t'_\alpha(\cos k_x + \cos k_y); \tag{2}$$

$$\varepsilon_k^{\alpha\alpha AB} = \varepsilon_k^{\alpha\alpha BA} = 4t_\alpha \cos\frac{k_x}{2}\cos\frac{k_y}{2}. \tag{3}$$

As mentioned in our main text, we can transfer the 4×4 TB model into a block-diagonalized matrix with using the glide symmetry:

$$H^{eff}(\boldsymbol{k}) = U H_t^{4-band} U^\dagger = \begin{pmatrix} H_t(\boldsymbol{k}) & 0 \\ 0 & H_t(\boldsymbol{k}+\boldsymbol{Q}) \end{pmatrix}, \tag{4}$$

here H_k is the effective two-band model in our main text and $Q = (\pi, \pi)$ is the folding vector. The unitary transformation U is defined as

$$U = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 \\ 1 & 0 & -1 & 0 \\ 0 & 1 & 0 & -1 \end{pmatrix}, \tag{5}$$

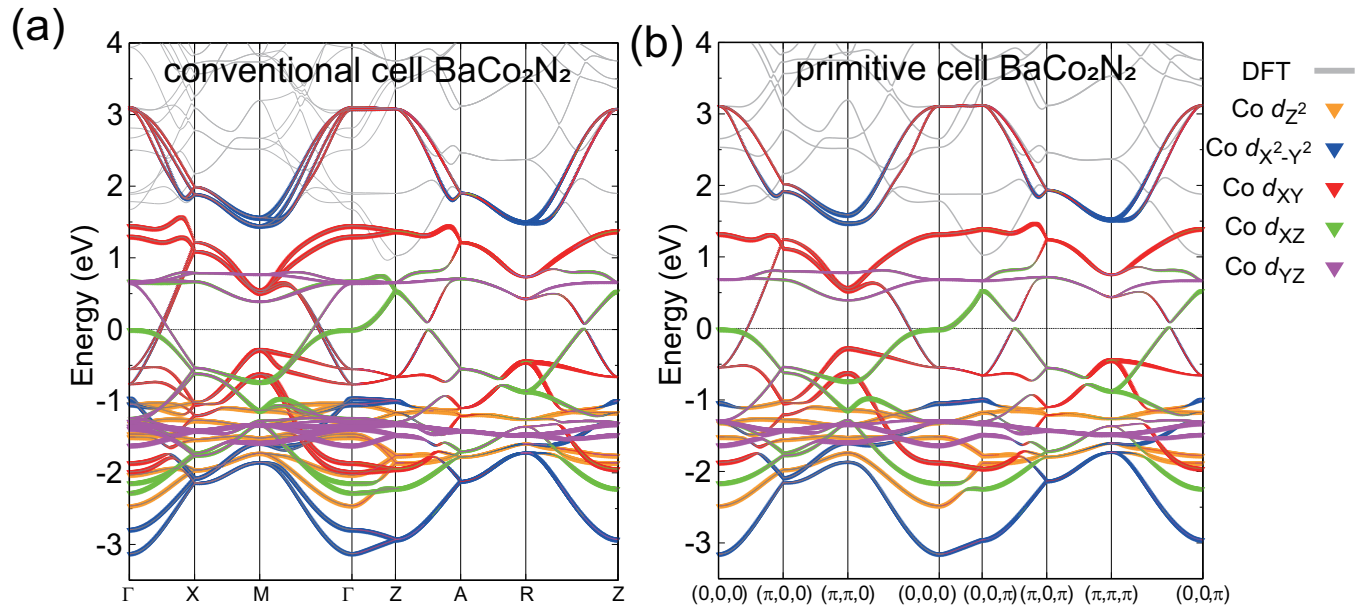

FIG. S1: DFT-calculated band structures of $BaCo_2N_2$ and projected weight of d-orbital-like WFs in its symmetric local coordinate (X-Y coordinate) with (a) the conventional cell and (b) the primitive cell. The coordinates in the abscissa of (b) are corresponding to the high symmetric **k**-points of the tetragonal conventional cell's Brillouin zone.

which is similar to that in FeSe[6] and $BaCuS_2$[7].

BAND STRUCTURE OF $BaCo_2N_2$

Fig.S1 shows the orbital-resolved band structure from DFT calculations of $BaCo_2N_2$. The N p orbitals, absent in the figure, are located around 3.5 eV below the Fermi level, while Co 3d orbitals dominate from -3 eV to 3 eV. Despite moderate inter-orbital coupling, low-energy bonding states and high-energy anti-bonding states for each Co 3d orbital can be identified. We also notice that the dispersion of band structure around the Fermi level is similar with that in Co_2N_2 monolayer, which is consistent with our previous qualitative analysis from symmetry.

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