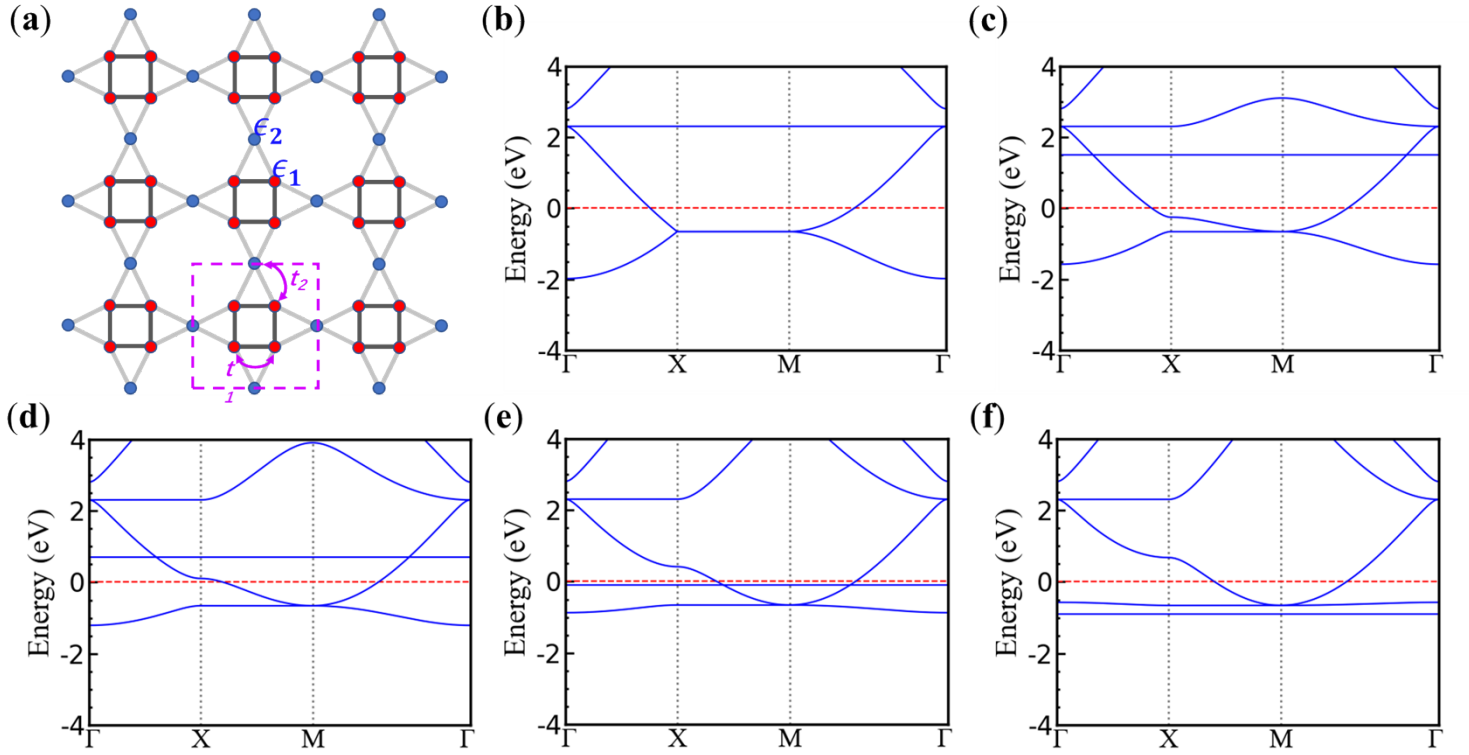


# Coupling double flat bands in a quadrangular-star lattice

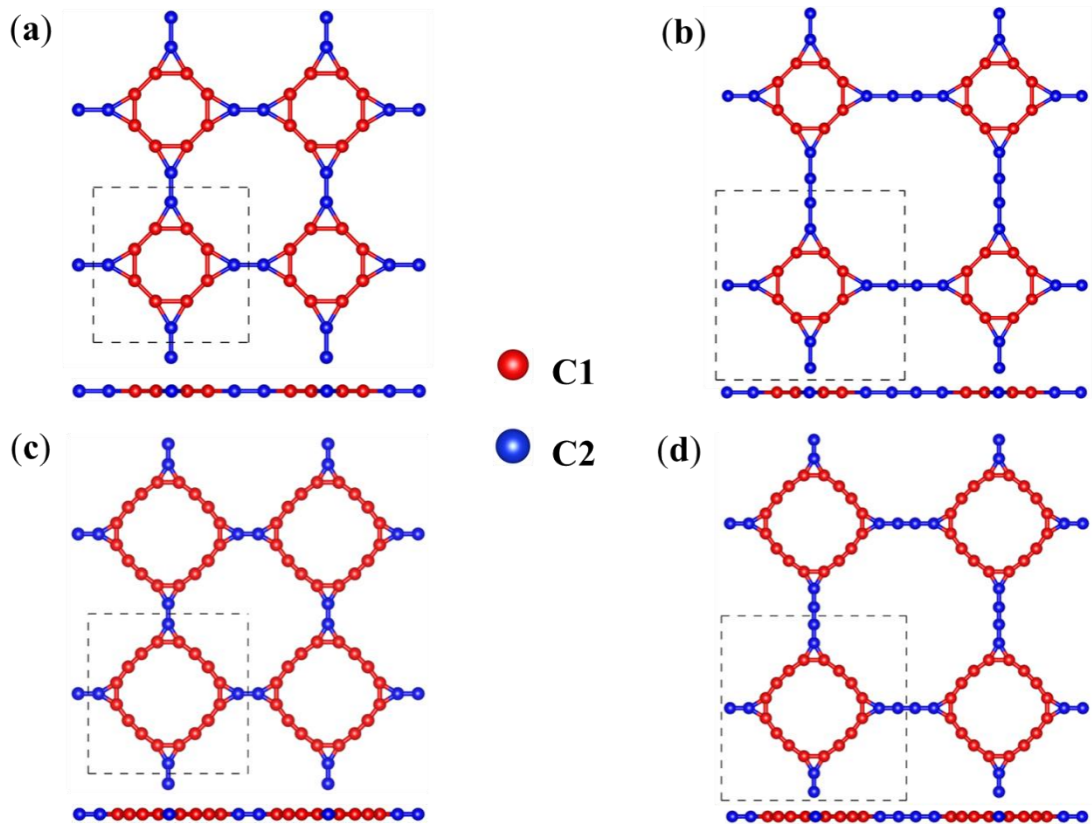
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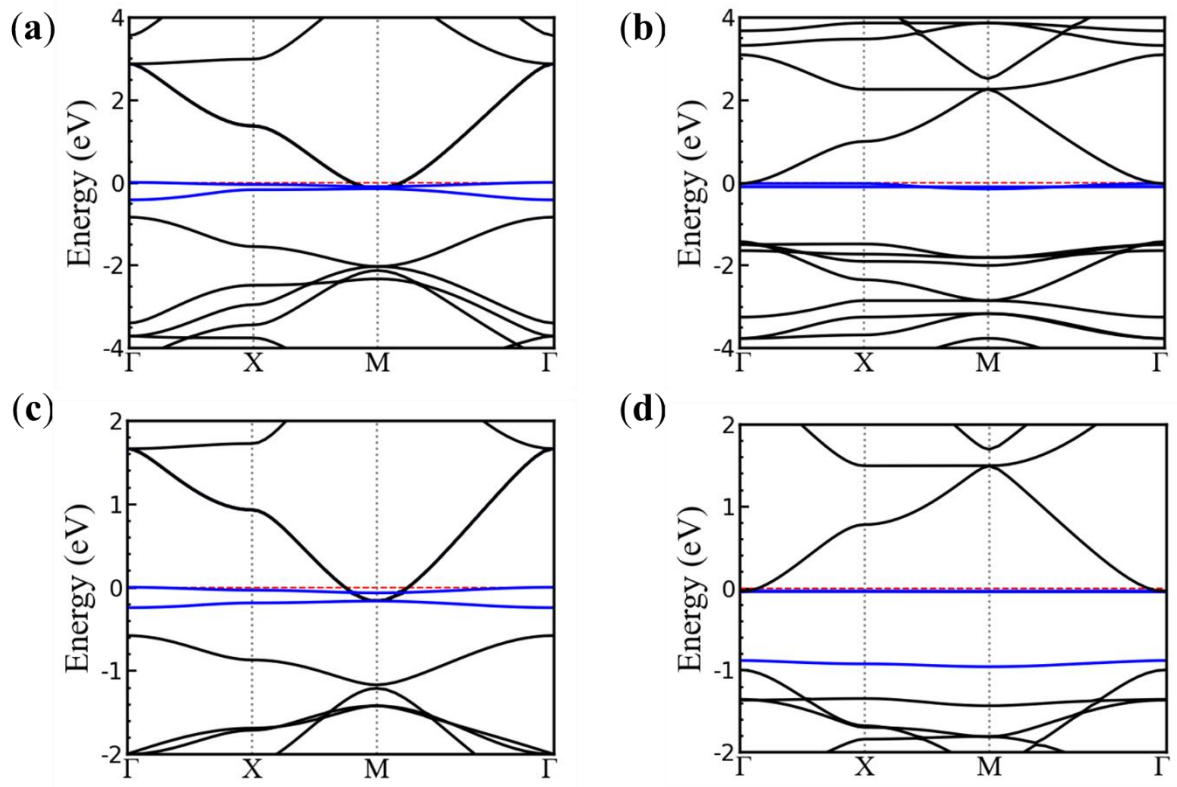
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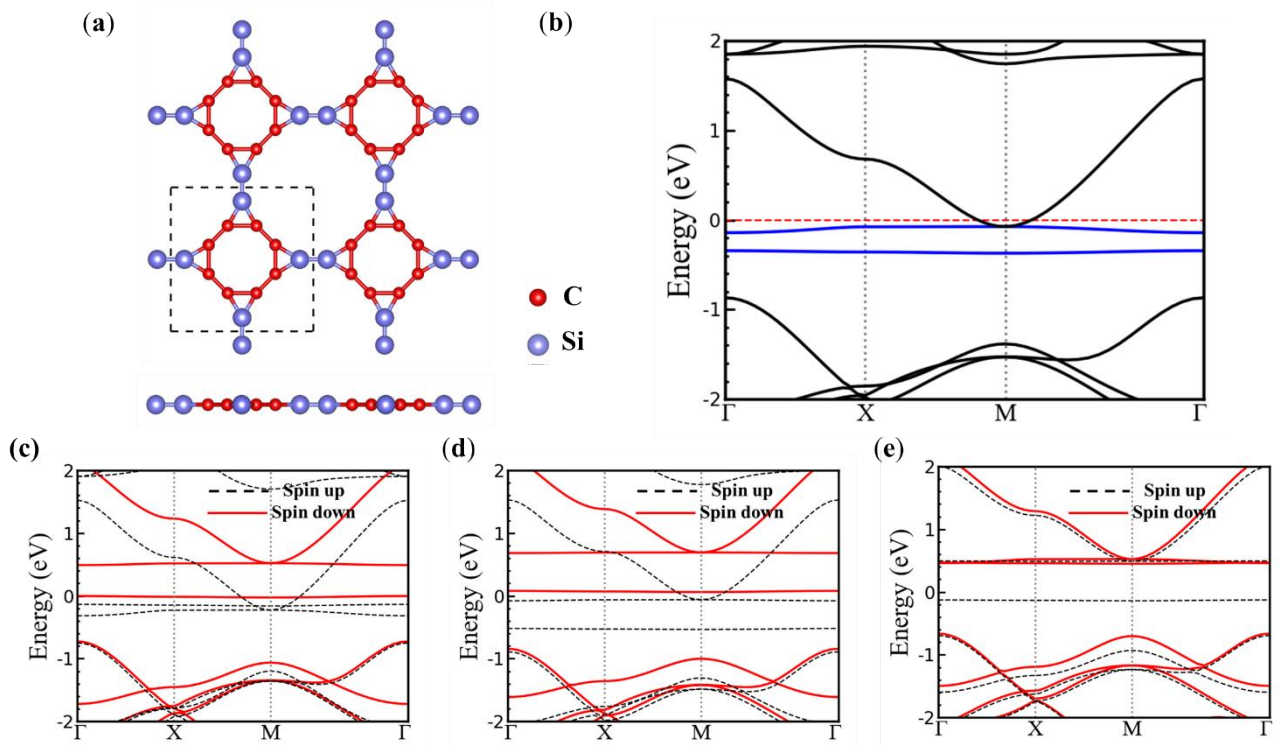
**FIG. S1.** (a) A QSL, in which the primitive unit cell is shown in the dotted box. The site energies of the red and blue lattices are marked as  $\epsilon_1$  and  $\epsilon_2$ ,  $t_1$  and  $t_2$  represent the hopping energies between red lattices and between red and blue lattices, respectively. Based on the band structure of tight-binding model in Eq. (1) with parameters  $\epsilon_1 = 2.8$ ,  $\epsilon_2 = 2.3$ , and (b)  $t_1=0$  eV,  $t_2=1.6$  eV; (c)  $t_1 = 0.4$  eV,  $t_2 = 1.6$  eV; (d)  $t_1 = 0.6$  eV,  $t_2 = 1.6$  eV; (e)  $t_1 = 1.2$  eV,  $t_2 = 1.6$  eV; (f)  $t_1 = 1.6$  eV,  $t_2 = 1.6$  eV.



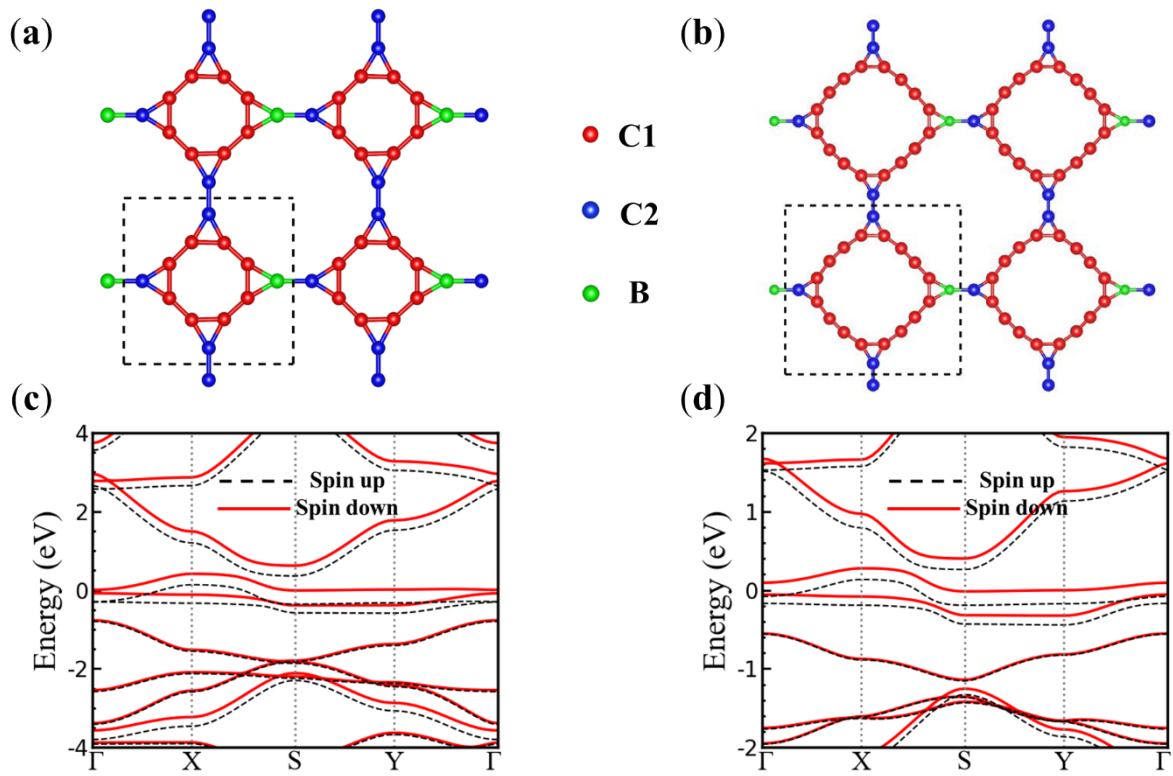
**FIG. S2.** The top view and side views of CQSL-12 (a), CQSL-16 (b), CQSL-20 (c) and CQSL-24 (d), where their primitive cells are shown in the dashed boxes.



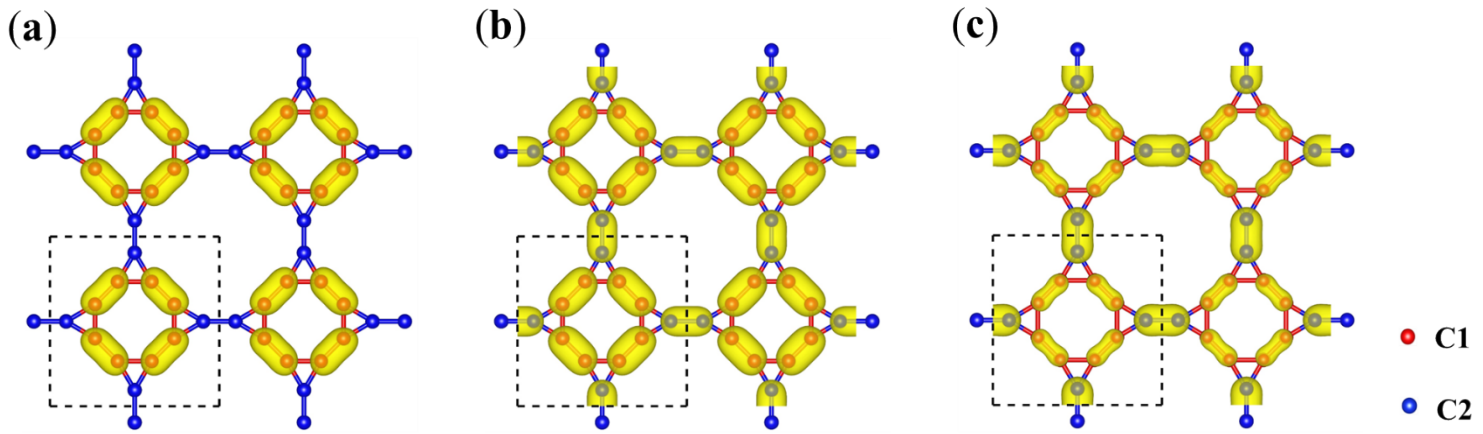
**FIG. S3.** Band structures of CQSL-12 (a), CQSL-16 (b), CQSL-20 (c) and CQSL-24 (d).



**FIG. S4.** (a) Atomic structure of CSiQSL-12, where the carbon dimers in CQSL-12 are replaced by silicon atoms. (b) Band structure of CSiQSL-12. Band structures of CSiQSL-12 in the case of one (c), two (d) and three (e) holes doping. The dashed and solid lines represent the spin-up and spin-down energy bands, respectively.



**FIG. S5.** Structure diagram of CBQSL-12 (a) and CBQSL-20 (b). The energy band structures of CBQSL-12 (c) and CBQSL-20 (d), where dotted and solid lines represent the spin up and spin down energy bands, respectively.



**FIG. S6.** The spin polarization density of QSL-12 doped with 1 (a), 2 (b), and 3 (c) holes. We set the isosurface level to  $0.003 \text{ e/bohr}^3$ .