## SUPPLEMENTAL MATERIALS: IMPLEMENTATION

The 3DSOC can be implemented in <sup>87</sup>Rb using two photon transitions. A possible implementation is given in Fig. ??. Nine laser beams with wavelength  $\lambda$  are used to couple states within the F = 1 and F = 2 hyperfine manifolds. A Zeeman field of B = 200mT sets the quantization axis along the  $\hat{z}$  direction. The remaining hyperfine transitions are isolated with a 6.8 GHz microwave field. Each pair of non-adjacent couplings is induced with three laser beams. All lasers have frequencies tuned between the  $D_1$  and  $D_2$  transitions. To ensure that all unwanted couplings are off resonance, we chose the base frequencies much larger than the splitting between the hyperfine levels  $\omega_a, \omega_b, \omega_c \gg \delta_{ij}$ .

The six optical couplings are induced in non-adjacent pairs on the state-linkage diagram. We list the properties of the three pairs of couplings independently. In what follows we define the vectors  $\hat{e}_{\pm} = \pm \frac{1}{\sqrt{2}} (\hat{x} \pm \hat{y})$  and  $\hbar k_L$ is the recoil momentum of the laser.

# Couplings $\Omega_{12}$ and $\Omega_{34}$

The two couplings  $\Omega_{12}$  and  $\Omega_{34}$  will be induced with three lasers, denoted  $L_a$ ,  $L_{12}$  and  $L_{34}$ . The beam  $L_a$  will be shared in the two-photon couplings. The frequencies of the three beams will be chosen such that

$$\omega_{12} = \omega_a + \delta_{12} \tag{1}$$

$$\omega_{34} = \omega_a + \delta_{34},\tag{2}$$

where  $\omega_a$  is the frequency of the laser  $L_a$  and  $\delta_{ij}$ , is the frequency splitting between the states  $|i\rangle$  and  $|j\rangle$ . With such a configuration, the transition  $|1\rangle \leftrightarrow |2\rangle$  and  $|3\rangle \leftrightarrow$  $|4\rangle$  will be on resonance, while all other dipole allowed transitions will be off resonance. The wavevectors of the lasers are given by

$$\boldsymbol{\kappa}_a = k_L \hat{z} \tag{3}$$

$$\boldsymbol{\kappa}_{12} = -k_L \hat{\boldsymbol{e}}_- \tag{4}$$

$$\boldsymbol{\kappa}_{34} = k_L \hat{\boldsymbol{e}}_{-}.\tag{5}$$

We can check that the effective momentum transfer of the couplings are given by

$$\mathbf{k}_{12} = k_L(\hat{e}_- + \hat{z}) \tag{6}$$

$$=\frac{k_L}{\sqrt{2}}\left(\hat{x}-\hat{y}+\sqrt{2}\hat{z}\right) \tag{7}$$

$$=\mathbf{K}_1 - \mathbf{K}_2 \tag{8}$$

and

$$\mathbf{k}_{34} = k_L(-\hat{e}_- + \hat{z}) \tag{9}$$

$$=\frac{k_L}{\sqrt{2}}\left(\hat{y}-\hat{x}+\sqrt{2}\hat{z}\right)\tag{10}$$

$$= \mathbf{K}_3 - \mathbf{K}_4. \tag{11}$$



 $m_F$ 

FIG. 1. Four hyperfine states  $|F, m_F\rangle$  of <sup>87</sup>Rb are coupled using nine lasers. The quantization axis is set by a Zeeman field along the  $\hat{z}$ -axis. The couplings are produced in pairs. (a) The four states in the tetrahedral coupling are mapped to physical states according to  $|1\rangle = |2, 0\rangle$ ,  $|2\rangle = |1, +1\rangle$ ,  $|3\rangle = |1, 0\rangle$ ,  $|1\rangle = |2, +1\rangle$ . (b) The frequencies of the three sets of lasers are given by  $\{\omega_a, \omega_a + \delta_{12}, \omega_a + \delta_{34}\}$  (dashed blue),  $\{\omega_b, \omega_b + \delta_{13}, \omega_b + \delta_{24}\}$  (dotted black), and  $\{\omega_c, \omega_c + \delta_{14}, \omega_c + \delta_{23}\}$  (solid red), where  $\delta_{ij} = \omega_i - \omega_j$  is the frequency difference between the states  $|i\rangle$  and  $|j\rangle$  in the rotating frame. (c) The geometry of the nine laser beams.  $-\mathbf{k}_{12} = -\mathbf{k}_{13} = \mathbf{k}_{23} =$  $\mathbf{k}_{34} = k_L \hat{e}_-$ ,  $\mathbf{k}_a = \mathbf{k}_{23} = -\mathbf{k}_{14} = k_L \hat{z}$  and  $\mathbf{k}_b = -\mathbf{k}_c = k_L \hat{e}_-$ . The unit vectors  $\hat{e}_{\pm} = \pm \frac{1}{\sqrt{2}}(\hat{x} \pm \hat{y})$ .

 $L_{a}L_{23}$ 

 $\hat{e}$ 

Finally, the polarizations of the lasers will be chosen such that the beams  $L_{12}$  and  $L_{34}$  are linearly polarized along the  $\hat{z}$  direction, while  $L_a$  is  $\sigma_+$  polarized.

# Couplings $\Omega_{14}$ and $\Omega_{23}$

The couplings  $\Omega_{14}$  and  $\Omega_{23}$  will similarly be induced with three lasers, denoted  $L_b$ ,  $L_{14}$  and  $L_{23}$ . The beam  $L_b$  will be shared in the two-photon couplings. The frequencies of the three beams will be chosen such that

$$\omega_{14} = \omega_b + \delta_{14} \tag{12}$$

$$\omega_{23} = \omega_b + \delta_{23},\tag{13}$$

where  $\omega_b$  is the frequency of the laser  $L_b$ . This choice of frequencies will isolate the transitions  $|1\rangle \leftrightarrow |4\rangle$  and  $|2\rangle \leftrightarrow |3\rangle$  in a manner similar to above couplings. The wavevectors of the lasers are given by

$$\boldsymbol{\kappa}_b = k_L \hat{\boldsymbol{e}}_+ \tag{14}$$

$$\boldsymbol{\kappa}_{14} = -k_L \hat{z} \tag{15}$$

$$\boldsymbol{\kappa}_{23} = k_L \hat{z}.\tag{16}$$

We can check that the effective momentum transfer of the couplings are given by

$$\mathbf{k}_{14} = k_L(\hat{e}_+ + \hat{z}) \tag{17}$$

$$=\frac{k_L}{\sqrt{2}}\left(\hat{x}+\hat{y}+\sqrt{2}\hat{z}\right)\tag{18}$$

$$=\mathbf{K}_1-\mathbf{K}_4\tag{19}$$

and

$$\mathbf{k}_{23} = k_L (\hat{e}_+ - \hat{z}) \tag{20}$$

$$= \frac{\kappa_L}{\sqrt{2}} \left( \hat{x} + \hat{y} - \sqrt{2}\hat{z} \right) \tag{21}$$

$$= \mathbf{K}_2 - \mathbf{K}_3. \tag{22}$$

Finally, the polarizations of the lasers will be chosen such that the beams  $L_b$  is linearly polarized along the  $\hat{z}$  direction, while  $L_{14}$  and  $L_{23}$  are linearly polarized along the  $\hat{x}$  axis.

#### Couplings $\Omega_{13}$ and $\Omega_{24}$

The last pair of couplings  $\Omega_{13}$  and  $\Omega_{24}$  will similarly be induced with three lasers, denoted  $L_c$ ,  $L_{13}$  and  $L_{24}$ . The beam  $L_c$  will be shared in the two-photon couplings. The frequencies of the three beams will be chosen such that

$$\omega_{13} = \omega_c + \delta_{13} \tag{23}$$

$$\omega_{24} = \omega_c + \delta_{24},\tag{24}$$

where  $\omega_c$  is the frequency of the laser  $L_c$ . This choice of frequencies will isolate the transitions  $|1\rangle \leftrightarrow |3\rangle$  and  $|2\rangle \leftrightarrow |4\rangle$  in a manner similar to above couplings. The wavevectors of the lasers are given by

$$\boldsymbol{\kappa}_c = -k_L \hat{\boldsymbol{e}}_+ \tag{25}$$

$$\boldsymbol{\kappa}_{13} = -k_L \hat{e}_- \tag{26}$$

$$\boldsymbol{\kappa}_{24} = k_L \hat{e}_-. \tag{27}$$

We can check that the effective momentum transfer of the couplings are given by

$$\mathbf{k}_{13} = k_L(-\hat{e}_+ + \hat{e}_-) \tag{28}$$

$$=\frac{k_L}{\sqrt{2}}\left(2\hat{x}\right)\tag{29}$$

$$=\mathbf{K}_1-\mathbf{K}_3\tag{30}$$

and

$$\mathbf{k}_{24} = k_L(-\hat{e}_+ - \hat{e}_-) \tag{31}$$

$$=\frac{k_L}{\sqrt{2}}\left(2\hat{y}\right)\tag{32}$$

$$\mathbf{K}_2 - \mathbf{K}_4. \tag{33}$$

Finally, the polarizations of the lasers will be chosen such that the beams  $L_{12}$  and  $L_{34}$  are linearly polarized along the  $\hat{e}_+$  direction, while  $L_c$  is linearly polarized along  $\hat{e}_-$ .

## Amplitude and Phase

In each pair of transitions, the three lasers provide a sufficient number of both amplitude and phase degrees of freedom to chose the the values of the couplings as desired in the main text.