The original Hamiltonian H_{couple} is constructed by

$$
\begin{aligned}
H_{couple} &= \begin{bmatrix} \omega_q + \omega_q' & 0 & g\cos\theta_2 & g\sin\theta_2 \\ 0 & \omega_q - \omega_q' & g\sin\theta_2 & -g\cos\theta_2 \\ g\cos\theta_2 & g\sin\theta_2 & -\omega_q + \omega_q' & 0 \\ g\sin\theta_2 & -g\cos\theta_2 & 0 & -\omega_q - \omega_q' \end{bmatrix} \\
&= \begin{bmatrix} \omega_q I & g\cos\theta_2 I \\ g\cos\theta_2 I & -\omega_q I \end{bmatrix} + \begin{bmatrix} \omega_q'\sigma_z & \begin{bmatrix} 0 & g\sin\theta_2 \\ g\sin\theta_2 & -2g\cos\theta_2 \end{bmatrix} \\ \begin{bmatrix} 0 & g\sin\theta_2 \\ g\sin\theta_2 & -2g\cos\theta_2 \end{bmatrix} & \omega_q'\sigma_z \end{bmatrix} \\
&= \begin{bmatrix} \omega_q & g\cos\theta_2 \\ g\cos\theta_2 & -\omega_q \end{bmatrix} \otimes I + \begin{bmatrix} \omega_q'\sigma_z & G \\ G & \omega_q'\sigma_z \end{bmatrix} \\
&= H_1 + H_2
\end{aligned} \tag{1}
$$

The diagonalized Hamiltonian is given by

$$
H_{diagonalized} = \begin{bmatrix} \lambda_1 & 0 & 0 & 0 \\ 0 & \lambda_2 & 0 & 0 \\ 0 & 0 & \lambda_3 & 0 \\ 0 & 0 & 0 & \lambda_4 \end{bmatrix} \tag{2}
$$

Where

$$
\begin{aligned}
\lambda_1 &= -\sqrt{(\omega_q' + \Omega)^2 + (g\sin\theta_2)^2} \\
\lambda_1 &= -\sqrt{(\omega_q' - \Omega)^2 + (g\sin\theta_2)^2} \\
\lambda_1 &= \sqrt{(\omega_q' - \Omega)^2 + (g\sin\theta_2)^2} \\
\lambda_1 &= \sqrt{(\omega_q' + \Omega)^2 + (g\sin\theta_2)^2} \\
\Omega &= \sqrt{\omega_q^2 + (g\cos\theta_2)^2}
\end{aligned} \tag{3}
$$

Here we construct three rotation matrices $R_1(\alpha)$, $R_2(\phi_1)$, and $R_3(\phi_2)$ which are defined by

$$
R_1(\alpha) = \begin{bmatrix} \cos\alpha & -\sin\alpha \\ \sin\alpha & \cos\alpha \end{bmatrix} \otimes I \tag{4}
$$

$$
R_2(\phi_1) = \begin{bmatrix} \cos\phi_1 & 0 & 0 & -\sin\phi_1 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ \sin\phi_1 & 0 & 0 & \cos\phi_1 \end{bmatrix} \tag{5}
$$

$$
R_3(\phi_2) = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos\phi_2 & -\sin\phi_2 & 0 \\ 0 & \sin\phi_2 & \cos\phi_2 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} \tag{6}
$$

Where

$$
\sin\alpha = \frac{1}{\sqrt{2}} \frac{g\cos\theta_2}{\sqrt{\Omega(\Omega + \omega_q')}} \tag{7}
$$

$$\cos\alpha = \frac{1}{\sqrt{2}}\sqrt{\frac{\Omega+\omega_q}{\Omega}} \tag{8}$$

$$\sin\phi_1 = \frac{1}{\sqrt{2}}[1-\frac{\omega_q'+\Omega}{(g\sin\theta_2)^2+(\omega_q'+\Omega)^2}]^{-\frac{1}{2}}\frac{g\sin\theta_2}{\sqrt{(g\sin\theta_2)^2+(\omega_q'+\Omega)^2}} \tag{9}$$

$$\cos\phi_1 = \frac{1}{\sqrt{2}}\sqrt{1-\frac{\omega_q'+\Omega}{(g\sin\theta_2)^2+(\omega_q'+\Omega)^2}} \tag{10}$$

$$\sin\phi_2 = \frac{1}{\sqrt{2}}[1-\frac{\Omega-\omega_q'}{(g\sin\theta_2)^2+(\omega_q'-\Omega)^2}]^{-\frac{1}{2}}\frac{g\sin\theta_2}{\sqrt{(g\sin\theta_2)^2+(\omega_q'-\Omega)^2}} \tag{11}$$

$$\cos\phi_2 = \frac{1}{\sqrt{2}}\sqrt{1-\frac{\Omega-\omega_q'}{(g\sin\theta_2)^2+(\omega_q'-\Omega)^2}} \tag{12}$$

And our unitary transformation operator is constructed by

$$\hat{U} = \hat{R}_3\hat{R}_2\hat{R}_1 = \begin{bmatrix} \cos\alpha\cos\phi_1 & \sin\alpha\sin\phi_2 & \sin\alpha\cos\phi_2 & -\cos\alpha\sin\phi_1 \\ -\sin\alpha\sin\phi_1 & \cos\alpha\cos\phi_2 & -\cos\alpha\sin\phi_2 & -\sin\alpha\cos\phi_1 \\ \sin\alpha\cos\phi_1 & -\cos\alpha\sin\phi_2 & -\cos\alpha\cos\phi_2 & -\sin\alpha\sin\phi_1 \\ -\cos\alpha\sin\phi_1 & -\sin\alpha\cos\phi_2 & \sin\alpha\sin\phi_2 & -\cos\alpha\cos\phi_1 \end{bmatrix} \tag{13}$$

Here is the derivation. Calculating $R_1 H_{couple} R_1^\dagger = R_1 H_1 R_1^\dagger + R_1 H_2 R_1^\dagger$

$$R_1 H_1 R_1^\dagger = \begin{bmatrix} \Omega & 0 \\ 0 & -\Omega \end{bmatrix} \otimes I \tag{14}$$

$$R_1 H_2 R_1^\dagger = \begin{bmatrix} \omega_q'\sigma_z - G\sin 2\alpha & G\cos 2\alpha \\ G\cos 2\alpha & \omega_q'\sigma_z + G\sin 2\alpha \end{bmatrix} \tag{15}$$

So, we get

$$\begin{aligned} R_1 H_{couple} R_1^\dagger &= \begin{bmatrix} \Omega I + \omega_q'\sigma_z - G\sin 2\alpha & G\cos 2\alpha \\ G\cos 2\alpha & -\Omega I + \omega_q'\sigma_z + G\sin 2\alpha \end{bmatrix} \\ &= \begin{bmatrix} \Omega I + \omega_q'\sigma_z & g\sin\theta_2 \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \\ g\sin\theta_2 \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} & -\Omega I + \omega_q'\sigma_z \end{bmatrix} + H_{residue} \\ &= H_{target} + H_{residue} \end{aligned} \tag{16}$$

Notice that

$$R_3 R_2 H_{target} R_2^\dagger R_3^\dagger = \begin{bmatrix} \lambda_1 & 0 & 0 & 0 \\ 0 & \lambda_2 & 0 & 0 \\ 0 & 0 & \lambda_3 & 0 \\ 0 & 0 & 0 & \lambda_4 \end{bmatrix} \tag{17}$$

So, we could deduce that

$$R_3 R_2 H_{residue} R_2^\dagger R_3^\dagger = 0 \tag{18}$$

Therefore, the unitary operator U is our target matrix which could diagonalize H_{couple}.