Supplemental Material for "Vanishing and Revival of Resonance Raman Scattering"

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I. ANALYTICAL SOLUTION FOR A FOUR-LEVEL DOUBLE-Λ ^{87}RB SYSTEM

We derive an analytic solution for a four-level double-Λ ^{87}Rb atom driven by a linearly polarized time-dependent laser pulse without using the rotating-wave approximation. The system consists of four states $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$ with energies E_1, E_2, E_3, and E_4 interacting with a pulsed laser field $\mathcal{E}(t)$. The corresponding Hamiltonian in the dipole approximation can be written as

$$\hat{H}(t) = \begin{pmatrix} E_1 & 0 & 0 & 0 \\ 0 & E_2 & 0 & 0 \\ 0 & 0 & E_3 & 0 \\ 0 & 0 & 0 & E_4 \end{pmatrix} - \begin{pmatrix} 0 & 0 & \mu_{13} & \mu_{14} \\ 0 & 0 & \mu_{23} & \mu_{24} \\ \mu_{13} & \mu_{23} & 0 & 0 \\ \mu_{14} & \mu_{24} & 0 & 0 \end{pmatrix} \mathcal{E}(t) \tag{S1}$$

where the dipole matrix elements satisfy the following relations

$$\begin{aligned} \mu_{14} &= \sqrt{\frac{1}{4}}\mu_J = -\sqrt{3}\mu_{13}, \\ \mu_{13} &= -\sqrt{\frac{1}{12}}\mu_J, \\ \mu_{23} &= \sqrt{\frac{1}{4}}\mu_J = \sqrt{3}\mu_{24}, \\ \mu_{24} &= \sqrt{\frac{1}{12}}\mu_J, \end{aligned} \tag{S2}$$

with μ_J the transition dipole matrix element of $5^2S_{1/2} \rightarrow 5^2P_{1/2}$ [1]. In the interaction picture, the Hamiltonian in Eq. (S1) can be rewritten as

$$\hat{H}_I = -\begin{pmatrix} 0 & 0 & \mu_{13}\mathcal{E}(t)\,e^{-i\omega_{13}t} & \mu_{14}\mathcal{E}(t)\,e^{-i\omega_{14}t} \\ 0 & 0 & \mu_{23}\mathcal{E}(t)\,e^{-i\omega_{23}t} & \mu_{24}\mathcal{E}(t)\,e^{-i\omega_{24}t} \\ \mu_{13}\mathcal{E}(t)\,e^{i\omega_{13}t} & \mu_{23}\mathcal{E}(t)\,e^{i\omega_{23}t} & 0 & 0 \\ \mu_{14}\mathcal{E}(t)\,e^{i\omega_{14}t} & \mu_{24}\mathcal{E}(t)\,e^{i\omega_{24}t} & 0 & 0 \end{pmatrix} \tag{S3}$$

with $\omega_{nm} = E_m - E_n$. In the broad-bandwidth-limit regime, i.e., $\Delta\omega \gg \delta_2$, the states $|3\rangle$ and $|4\rangle$ can be regarded as near degenerate in energy. To this end, we consider the limit case when $\omega_{14} = \omega_{13}$ and $\omega_{24} = \omega_{23}$, and therefore Eq. (S3) can be written as

$$\hat{H}_I = -\begin{pmatrix} 0 & 0 & \mu_{13}\mathcal{E}(t)\,e^{-i\omega_{13}t} & \mu_{14}\mathcal{E}(t)\,e^{-i\omega_{13}t} \\ 0 & 0 & \mu_{23}\mathcal{E}(t)\,e^{-i\omega_{23}t} & \mu_{24}\mathcal{E}(t)\,e^{-i\omega_{23}t} \\ \mu_{13}\mathcal{E}(t)\,e^{i\omega_{13}t} & \mu_{23}\mathcal{E}(t)\,e^{i\omega_{23}t} & 0 & 0 \\ \mu_{14}\mathcal{E}(t)\,e^{i\omega_{13}t} & \mu_{24}\mathcal{E}(t)\,e^{i\omega_{23}t} & 0 & 0 \end{pmatrix}. \tag{S4}$$

By using the Magnus expansion [2], the time-evolution of the unitary operator can be written as

$$U(t,t_0) = \exp\left[\sum_{n=1}^{\infty} S^{(n)}(t)\right]. \tag{S5}$$

The first leading term is $S^{(1)}(t) = iA(t)$, with

$$\begin{aligned} A(t) &= -\int_{t_0}^{t} H_I(t_1)\, dt_1 \\ &= \begin{pmatrix} 0 & 0 & \theta_{13}^* & -\sqrt{3}\theta_{13}^* \\ 0 & 0 & \sqrt{3}\theta_{24}^* & \theta_{24}^* \\ \theta_1 & \sqrt{3}\theta_{24} & 0 & 0 \\ -\sqrt{3}\theta_{13} & \theta_{24} & 0 & 0 \end{pmatrix} \end{aligned} \tag{S6}$$

where

$$\theta_{13}(t) = \int_{t_0}^{t} \mu_{13}\mathcal{E}(t')\, e^{i\omega_{13}t'} dt, \tag{S7}$$

$$\theta_{24}(t) = \int_{t_0}^{t} \mu_{24}\mathcal{E}(t')\, e^{i\omega_{23}t'} dt. \tag{S8}$$

The corresponding unitary operator, in terms of eigenvalues and eigenvectors of $A(t)$, can be given as

$$\begin{aligned} U^{(1)}(t,t_0) &= \exp[iA(t)] \\ &= \sum_{n=1}^{4} \exp[iA_n(t)]|A_n\rangle\langle A_n| \end{aligned} \tag{S9}$$

where the eigenvalues A_n read

$$A_1 = -2\,|\theta_{13}|\,, \tag{S10}$$

$$A_2 = 2\,|\theta_{13}|\,, \tag{S11}$$

$$A_3 = -2\,|\theta_{24}|\,, \tag{S12}$$

$$A_4 = 2\,|\theta_{24}|\,, \tag{S13}$$

and the corresponding eigenvectors $|A_n\rangle$ are

$$|A_1\rangle = \frac{\theta_{13}^*}{\sqrt{2}\,|\theta_{13}|}\,|1\rangle - \frac{1}{2\sqrt{2}}\,|3\rangle + \sqrt{\frac{3}{8}}\,|4\rangle\,, \tag{S14}$$

$$|A_2\rangle = -\frac{\theta_{13}^*}{\sqrt{2}\,|\theta_{13}|}\,|1\rangle - \frac{1}{2\sqrt{2}}\,|3\rangle + \sqrt{\frac{3}{8}}\,|4\rangle\,, \tag{S15}$$

$$|A_3\rangle = -\frac{\theta_{24}^*}{\sqrt{2}\,|\theta_{24}|}\,|2\rangle + \sqrt{\frac{3}{8}}\,|3\rangle + \frac{1}{2\sqrt{2}}\,|4\rangle\,, \tag{S16}$$

$$|A_4\rangle = \frac{\theta_{24}^*}{\sqrt{2}\,|\theta_{24}|}\,|2\rangle + \sqrt{\frac{3}{8}}\,|3\rangle + \frac{1}{2\sqrt{2}}\,|4\rangle\,. \tag{S17}$$

For the system initially in $|1\rangle$ at $t = t_0$, the time-dependent wave function $|\psi^{(1)}(t) = U^{(1)}(t, t_0)|1\rangle$ for the four-level system can be obtained in terms of the complex pulse area by

$$|\psi^{(1)}(t)\rangle = \cos[\theta(t)]|1\rangle + \frac{i\theta_1(t)}{2\theta(t)}\sin[\theta(t)]|3\rangle + \frac{i\sqrt{3}\theta_1(t)}{2\theta(t)}\sin[\theta(t)]|4\rangle \tag{S18}$$

with $\theta_1(t) = 2\theta_{13}(t)$, and $\theta(t) = |\theta_1(t)|$. As a result, the four-level system is reduced to a three-level V system without the population in state $|2\rangle$ at any time t. This fact has been demonstrated in Fig. S1 by calculating the time-dependent populations of states $P_n(t) = |\langle n|\psi(t)|^2$, $(n = 1, 2, 3, 4)$, for the four-level double-Λ system, which is driven by using a pulsed laser field with a broad bandwidth of $\Delta\omega = 23\delta_1$. That is, the RRS contribution to the state $|2\rangle$ is annihilated in real time.

II. PULSE AREA THEOREM FOR A THREE-LEVEL V SYSTEM

As is evident from Eq. (S18), the final populations in the excited states depend on the pulse area of $\theta_{13}(t_f)$, i.e.,

$$P_3^{(1)}(t_f) = \left|\frac{i\theta_1(t_f)}{2\theta(t_f)}\sin[\theta(t_f)]\right|^2, \tag{S19}$$

and

$$P_4^{(1)}(t_f) = \left|\frac{i\sqrt{3}\theta_1(t_f)}{2\theta(t_f)}\sin[\theta(t_f)]\right|^2. \tag{S20}$$

A pulse area of $\theta_{13}(t_f) = \pi/4$ will lead to a population distribution of 1:3 with 25% in $|3\rangle$ and 75% in $|4\rangle$. To achieve this pulse area, we take the pulsed laser field as

$$\mathcal{E}(t) = \mathrm{Re}\left[\frac{1}{2\pi}\int_0^\infty A(\omega)e^{-i\omega t}e^{i\phi(\omega)}d\omega\right] \tag{S21}$$

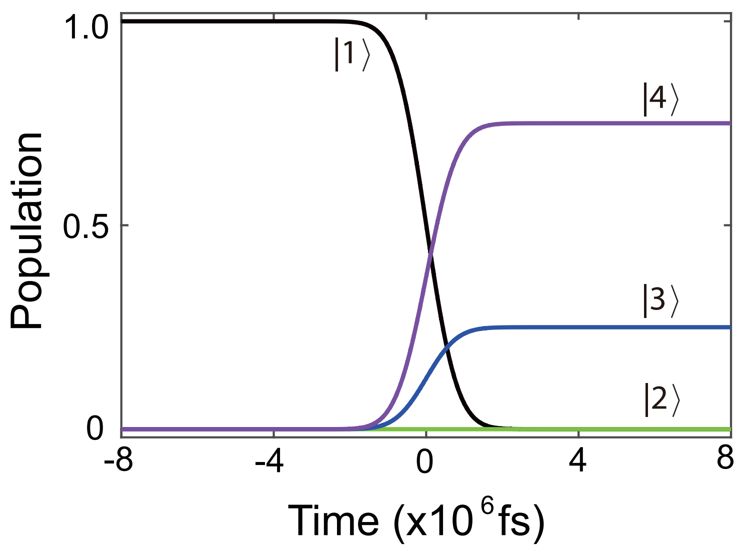

FIG. S1. The time-dependent populations of the states $P_n(t) = |\langle n|\psi(t)\rangle|^2$, $(n = 1, 2, 3, 4)$, for the four-level double-Λ system.

with the spectral amplitude

$$A(\omega) = \frac{A_0}{\mu_{13}} \exp\left[-\frac{(\omega - \omega_0)^2}{2(\Delta\omega)^2}\right]. \tag{S22}$$

At the resonant condition of $\omega_0 = \omega_{13}$, we obtain $\theta_{13}(t_f) = A(\omega_{13}) = A_0$. Therefore we fix $A_0 = \pi/4$ in our simulations. As can be seen from Fig. S1, the final populations $P_3(t_f)$ and $P_4(t_f)$ are in good agreement with this pulse area theorem by Eqs. (S19, S20)

[1] D. A. Steck, Rubidium 87 D Line Data, http://steck.us/alkalidata/rubidium87numbers.pdf.

[2] S. Blanes, F. Casas, J. A. Oteo, and J. Ros, The Magnus expansion and some of its applications, Phys. Rep. **470**, 151 (2009).