

Coherent population accumulations of multiphoton transitions induced by an ultrashort pulse train in polar molecules

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Coherent population accumulations of multiphoton transitions induced by an ultrashort pulse train in a two-level polar molecule are investigated theoretically by solving the density-matrix equations without invoking any of the standard approximations. It is shown due to the effects of permanent dipole moments, that the population accumulation of multiphoton transitions can be obtained in the polar molecule. Moreover, the population accumulations depend crucially on the relative phase between two sequential pulses, and the period in which the maximum population accumulation occurs is $2\pi/N$ in N -photon transitions.

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I. INTRODUCTION

In recent years, coherent control of multiphoton processes in atomic or molecular systems has been extensively investigated [1–7]. Most theoretical works on multiphoton excitations assumed that atoms or molecules do not possess permanent dipole moments. In fact, permanent dipole moments do exist in some quantum systems, such as polar molecules [8–11] and some solid materials [12–16], and must be considered in these dipole systems. In particular, the properties on the multiphoton resonance profiles can be greatly influenced in polar molecules [17]. As had been shown previously in one-color continuous wave (CW) [18], two-color CW [19], one-color pulsed [20,21], and two-color pulsed [22,23] when the dipole moment operators are nonzero, the selection rules and the molecule-laser excitations can be greatly modified [22]. In our previous work, we demonstrated that the corresponding physical picture of multiphoton excitations in dipole systems can be illustrated in an adiabatic interpretation [24]. Due to the effects of permanent dipole moments, the adiabatic eigenenergy difference is decreased when the level crossings occur, which changes the population transfer and enhances the multiphoton excitations.

Typical pulse-train effects, such as an accumulation of population and coherence, have attracted some researchers' attention [25–28]. If the laser repetition period is comparable with or smaller than the relaxation time of the system, the atomic systems will evolve as a result of the cumulative effect of these pulses [29,30]. The single-photon accumulative effects in two-level atoms for arbitrary pulse areas were investigated by an analytical theory which well agreed with the numerical results [28]. For accumulative effects involving two-photon absorption process, investigations are limited to three-level atomic system in which a substantial intermediate state is considered [31–33]. As for coherent population accumulation of multiphoton transitions in two-level media, es-

pecially in polar molecules, to our knowledge, it has not been investigated.

The general models for the train of pulses emerging from a mode-locked laser are that the time interval between two consecutive pulses is much longer than the pulse duration. Nevertheless, a necessary condition for manipulating some quantum coherence processes is the use of sufficiently short interacting times or short time intervals between pulses so that they can interact with the quantum system before it can be affected by its environment [34]. For instance, in order to repetitively drive selected vibrations of a molecular crystal lattice, a high-repetition-rate timed sequence of femtosecond pulses is applied [35]. Recently, a generation of a stable ultrahigh-repetition-rate train with hyperbolic secant envelopes has been demonstrated in an experiment [36], which can open many attractive applications in quantum coherent control.

In the case of a sequence of pulses, the relative phase of the two sequential laser pulses adds an additional degree of freedom. Investigations showed that the relative phase of picosecond, high-power nonoverlapping pulses can be adjusted by interferometric means, if the pulses are generated from the injection-locked system which forces pure amplitude modulation and leaves a collinear continuous laser beam, and the fluorescence suppression by π phase shifts can be demonstrated in such systems [37]. In the ultrashort pulse scheme, phase coherence among different transition pathways can produce interference effects on the resonantly enhanced transition probability in atomic systems [38]. The dispersion and reshaping effects for propagating in a resonant atomic medium were also shown to rely sensitively on the relative phase shift between the pulses of an ultrashort pulse train [39].

In this work, we apply an ultrashort pulse train to induce the coherent population accumulations of multiphoton transitions in a two-level polar molecule. It is demonstrated that the quantum interference between two sequential pulses can be enhanced because of the effects of permanent dipole moments, and the coherent population accumulation of multiphoton transitions, which cannot occur in nondipole systems, can be presented in the polar molecule. Moreover, the popu-

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lation accumulations depend crucially on the relative phase between the pulses, and the maximum population accumulation occurs in a relative phase period $2\pi/N$ in N -photon transitions.

This paper is organized as follows. The two-level polar molecule interacting with a femtosecond pulse train is described in Sec. II. The characteristics of the population accumulations induced by multiphoton transitions and the phase effects of the population accumulations are presented in Sec. III. We summarize our results and conclude in Sec. IV. Finally, an analytic process of a rotating wave approximation (RWA) to help interpret the exact results is shown in the Appendix.

II. THEORY

Consider a two-level polar molecule where $|1\rangle$ and $|2\rangle$ represent the ground and the excited states, with corresponding energies $E_1 = \frac{1}{2}\hbar\omega_0$ and $E_2 = \frac{3}{2}\hbar\omega_0$, respectively. ω_0 is the transition frequency between the two levels.

The electric field for the pulse train is

$$E(t) = \sum_0^n E_0 f(t - nT_R) \cos[\omega_l(t - nT_R) + n\phi], \quad (1)$$

where E_0 , ω_l , T_R , and ϕ are the electric field amplitude, the carrier circular frequency, the repetition period, and the relative phase between each pulse, respectively. The pulse temporal wave form $f(t - nT_R)$ is described as a realistic hyperbolic secant envelope [36]:

$$f(t - nT_R) = \text{sech}[(t - nT_R)/\tau_p]. \quad (2)$$

Here, τ_p is the duration of pulse. The total Hamiltonian describing the interaction of the field with the polar molecule is given by

$$\underline{H} = \underline{H}_0 + \underline{V} = \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix} - \underline{E}(t) \begin{bmatrix} \underline{\mu}_{11} & \underline{\mu}_{12} \\ \underline{\mu}_{21} & \underline{\mu}_{22} \end{bmatrix}. \quad (3)$$

Here $\underline{\mu}_{ij}$ are the dipole moment matrix elements. To simplify the problem, we assume that the laser field is linearly polarized, and the transition moment $\underline{\mu}_{21}$ and permanent moments $\underline{\mu}_{ij}$ are taken to be aligned with the direction of the polarization of the laser field.

The time evolution of the system is governed by the usual density-matrix equation

$$\frac{\partial \tilde{\rho}}{\partial t} = -\frac{i}{\hbar} [\tilde{H}, \tilde{\rho}]. \quad (4)$$

It is easy to derive the equations of the time-dependent ρ_{21} , ρ_{22} , and ρ_{11} as follows:

$$\frac{\partial \rho_{21}}{\partial t} = -i \left[\omega_0 \rho_{21} + (\mu_{21} \rho_{22} - \mu_{21} \rho_{11} - d \rho_{21}) \frac{E(t)}{\hbar} \right], \quad (5)$$

$$\frac{\partial \rho_{22}}{\partial t} = \frac{i}{\hbar} (\rho_{12} - \rho_{21}) \mu_{21} E(t), \quad (6)$$

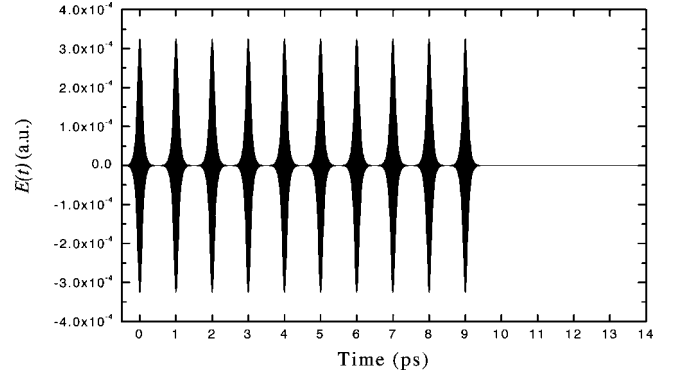


FIG. 1. The electric field of a sequence of $n=10$ pulses. The time parameters of the pulse train are $T_R=1.00$ ps and $\tau_p=1.00 \times 10^{-1}$ ps. The electric field amplitude of a laser pulse $E_0=3.25 \times 10^{-4}$ a.u. corresponds to a peak intensity of 3.71×10^9 W/cm².

$$\frac{\partial \rho_{11}}{\partial t} = \frac{i}{\hbar} (\rho_{21} - \rho_{12}) \mu_{21} E(t), \quad (7)$$

where $d = \mu_{22} - \mu_{11}$ is the difference in the permanent dipole moments between the ground and the excited levels. $dE(t)$ represents the effect of quantum interference resulting from the effects of permanent dipole moments. We should note for two-level polar molecules, that the enhancement of multiphoton transitions can be achieved even without the substantial intermediate states, which cannot occur in nondipole systems. With proper relative phase, population accumulation induced by multiphoton transitions can be obtained in polar molecules. The difference between permanent dipole moments d plays a very important role in the multiphoton transitions and the population accumulations as we will show later.

III. RESULTS AND DISCUSSION

The system of evolution Eqs. (5)–(7) is solved numerically with a fourth-order Runge-Kutta method. The RWA and the slowly varying envelope approximation (SVEA) are not employed in our numerical calculations.

The numerical simulations are based on molecular parameters characteristic of the $S_0 \rightarrow S_1$ electronic transition in 1-[*p*-(*N*, *N*-dimethylamino) phenyl]-4-(*p*-nitrophenyl)-1,3-butadiene which was involved in earlier studies [21,40] of the effects of permanent dipole moments on pulse interactions. In atomic units, the molecular parameters are $\omega_0=8.59 \times 10^{-2}$, $\mu_{21}=3.93$, and $d=1.18 \times 10$. The ultrashort pulse train is shown in Fig. 1. The time parameters of the pulse train are $T_R=1.00$ ps and $\tau_p=1.00 \times 10^{-1}$ ps, and the electric field amplitude of a laser pulse is $E_0=3.25 \times 10^{-4}$ a.u. corresponding to a peak intensity of 3.71×10^9 W/cm².

Figure 2 shows the time-dependent population behaviors of two-photon transitions ($\omega_l = \omega_0/2$) in the interaction of a molecule with the ultrashort pulse train of Fig. 1. In order to investigate the effects of permanent dipole moments on the population accumulations, we assume that there exists a

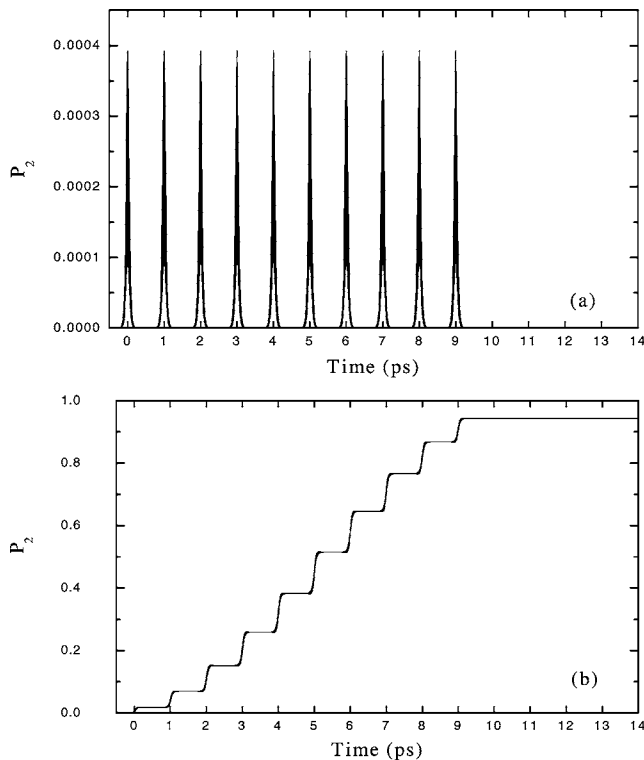


FIG. 2. The time-dependent population behaviors of two-photon transitions ($\omega_l = \omega_0/2$) in the interaction of a molecule with the ultrashort pulse train of Fig. 1. The molecule parameters are $\omega_0 = 8.59 \times 10^{-2}$ a.u. and $\mu_{21} = 3.93$ a.u.. (a) a pseudomolecule $d=0$ and (b) a realistic polar molecule $d=1.18 \times 10$ a.u..

pseudomolecule with the same parameters as the above molecular parameters except for $d=0$. The temporal population of excited state in the pseudomolecule is shown in Fig. 2(a). It can be found that two-photon transitions can hardly occur and there is no population accumulation in this case. However, when the effects of permanent dipole moments in the realistic polar molecule ($d=1.18 \times 10$ a.u.) are considered, the situation can be very different. It can be seen in Fig. 2(b), due to the presence of permanent dipole moments, that population accumulation induced by two-photon transitions can occur. Though a single-pulse-molecule interaction can lead to a relative small population transfer, the interference effects between two sequential pulses are greatly enhanced due to the effects of permanent dipole moments, which can result in population accumulation of two-photon transitions. Therefore, the maximum population of excited states can reach to a very high value (94.3%). The physical mechanism of the above phenomena can be understood as following. First, the paths of two-photon transitions can be opened because of the mixed parity in at least one of the two levels in the polar molecules [11,22]. When the situation is far away from single-photon resonance, the two-photon or multiphoton transitions become the most important factor determining population transfer. Moreover, the effects of permanent dipole moments modified the quantum coherence of interactions of polar molecules with the pulses [24], which can enlarge the interference effects between two sequential pulses. Therefore, population accumulations induced by mul-

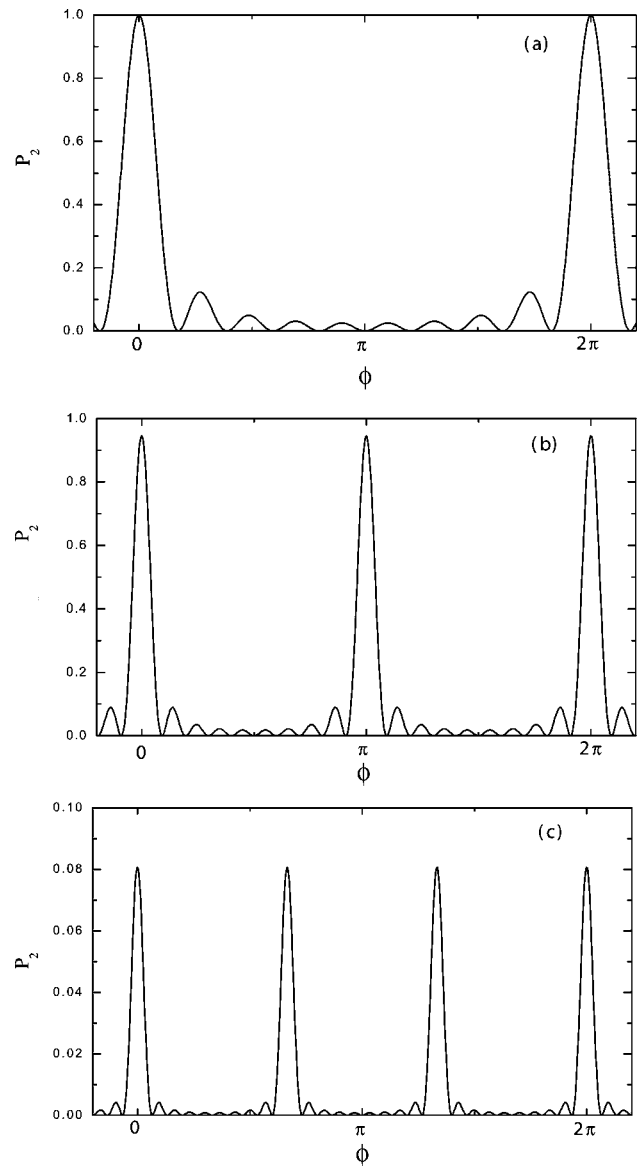


FIG. 3. Plot of the population accumulations in the excited state of the polar molecule as a function of the relative phase between two sequential pulses. The time parameters of the ultrashort pulse train are the same with those of Fig. 1. (a) $\omega_l = \omega_0$ and $E_0 = 1.08 \times 10^{-5}$ a.u., (b) $\omega_l = \omega_0/2$ and $E_0 = 3.25 \times 10^{-4}$ a.u., and (c) $\omega_l = \omega_0/3$ and $E_0 = 5.85 \times 10^{-4}$ a.u.

tiphoton transitions can occur in polar molecules.

In fact, the population accumulations of multiphoton transitions can be greatly affected by the relative phase between pulses. Figure 3 shows the population accumulations in the excited state of the polar molecule as a function of the relative phase between pulses ϕ . Figure 3(a) shows the population accumulations with the condition of a single-photon resonance ($\omega_l = \omega_0$). A relative small field strength $E_0 = 1.08 \times 10^{-5}$ a.u. is adopted. The maximal population accumulations occur when $\phi=0$ and $\phi=2\pi$. The further calculation shows that the population accumulation is a period function of the relative phase between pulses, and the period is 2π . In Fig. 3(b), the condition of a two-photon resonance ($\omega_l = \omega_0/2$) is satisfied. It can be seen that the maximum

population can be 94.3%, and the relative phase period of the population accumulation is $2\pi/2$. When three-photon resonance is satisfied, the field strength ($E_0=5.85 \times 10^{-4}$ a.u.) is increased due to the small transfer probability. The relative phase period is $2\pi/3$ which is shown in Fig. 3(c). The results can, of course, be obtained for other combinations of polar molecules and pulse trains.

In order to test the validity of the numerical results described above, we derived an analytic expression based on the RWA (The deducing process is shown in the Appendix.) In an interaction representation, the diagonal matrix elements of the Hamiltonian equal to zero and the off-diagonal matrix elements are given as following:

$$H_{12} = -\frac{\mu_{21}}{d}(n+1)N\omega_l \overbrace{J_N[zf(t)] \cdots J_N[zf(t-nT_R)]}^{n+1} \times \exp[inN\omega_l t] \overbrace{\exp[-iNT_R] \cdots \exp[-inNT_R]}^n \times \overbrace{\exp[inN\phi] \cdots \exp[inN\phi]}^n. \quad (8)$$

All the exponential terms under the last curly bracket in Eq. (8) tell the relationship between the interaction and the relative phase. It can be easily understood that H_{12} is a period function of ϕ . In other words, the interference effect is a period function of the relative phase between pulses. Moreover, it can be seen from Eq. (8) that the period in which the maximum population accumulation occurs is $2\pi/N$ in N -photon transitions. All the above analyses are consistent with our numerical results.

IV. CONCLUSIONS

In summary, we have analyzed the coherent population accumulations of multiphoton transitions induced by an ultrashort train in a polar molecule. Comparing with the pseudomolecule ($d=0$), the numerical results demonstrated due to the effects of permanent dipole moments, that the interference between pulses can be much stronger and the enhancement of population accumulation induced by multiphoton transitions can be obtained in the polar molecule ($d \neq 0$). Moreover, the population accumulations in the polar molecule depend sensitively to the relative phase between two sequential laser pulses, and the period in which the maximum population accumulation occurs is $2\pi/N$ in N -photon transitions. Furthermore, an analytic expression based on the RWA was deduced, which has confirmed the validity of the numerical results. The procedure developed in this work is to achieve the coherent control of the multiphoton transitions in a low intensity scheme, which might be helpful to control molecular-state populations and molecule-laser coupling in chemical reactions.

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APPENDIX: THE DEDUCING PROCESS OF THE ANALYTIC EXPRESSION

To locate the resonances in the transition probabilities, Eq. (4) is transferred into an interaction representation defined by

$$C_j = b_j \exp \left[\frac{i}{2} (-1)^{j+1} \left(\omega_0 t - d \int_0^t E(t') dt' \right) \right]. \quad (A1)$$

The diagonal matrix elements of the Hamiltonian equal to zero and the off-diagonal matrix elements are given by [18,23]

$$H_{12} = H_{21}^* = -\mu_{21} E(t) \exp \left[-i \left(\omega_0 t - d \int_0^t E(t') dt' \right) \right]. \quad (A2)$$

The crucial part of the above equation is the quantity

$$I = \exp[idE_0 Q], \quad (A3)$$

where

$$Q = \int_0^t f(t') \cos(\omega_l t' + \phi) dt'. \quad (A4)$$

Integrating Eq. (A4) by parts, one can obtain

$$Q = \frac{1}{\omega_l} f(t) \sin(\omega_l t + \phi) - \frac{1}{\omega_l} \int_0^t \frac{df(t')}{dt'} \sin(\omega_l t' + \phi) dt'. \quad (A5)$$

If the SVEA is satisfied, the second term in Eq. (A5) can be neglected, i.e.,

$$I = \exp[izf(t) \sin(\omega_l t + \phi)]. \quad (A6)$$

Here

$$z = \frac{dE_0}{\omega_l}. \quad (A7)$$

The ultrashort pulse train is adopted in our scheme, so that the off-diagonal matrix elements given by

$$H_{12} = -\mu_{21} E(t) \exp[-i\hbar\omega_0 t] \exp[izf(t) \sin \omega_l t] \times \exp\{izf(t-T_R) \sin[\omega_l(t-T_R) + \phi]\} \times \exp\{izf(t-2T_R) \sin[\omega_l(t-2T_R) + 2\phi]\} \cdots \times \exp\{izf(t-nT_R) \sin[\omega_l(t-nT_R) + n\phi]\}. \quad (A8)$$

Using the identity [23,41]

$$\exp[ix \sin \theta] = \sum_{k=-\infty}^{\infty} J_k(x) \exp[ik\theta], \quad (A9)$$

one can get

$$\begin{aligned}
 H_{12} = & -\mu_{21} E(t) \exp[-i\omega_0 t] \sum_{k=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \cdots \sum_{j=-\infty}^{\infty} \\
 & \times \overbrace{\{J_k[zf(t)] \exp[ik\omega_l t]\} \{J_m[zf(t-T_R)] \exp[im(\omega_l t + \phi)]\}}^{n+1} \\
 & \cdots \{J_j[zf(t-nT_R)]\} \exp\{ij[\omega_l(t-nT_R) + n\phi]\}.
 \end{aligned} \tag{A10}$$

It follows readily from Eq. (1) that

$$\begin{aligned}
 E(t) = & \sum_0^n \frac{1}{2} E_0 f(t-nT_R) (\exp\{i[\omega_l(t-nT_R) + n\phi]\} \\
 & + \exp\{-i[\omega_l(t-nT_R) + n\phi]\}).
 \end{aligned} \tag{A11}$$

On substituting for $E(t)$ from the above equation into Eq. (A10), one can obtain a sum equation with $(n+1)$ terms. One of these terms is

$$\begin{aligned}
 \text{term}(n) = & -\frac{1}{2} \mu_{21} E_0 \exp[-i\omega_0 t] f(t-nT_R) \\
 & \times \sum_{j=-\infty}^{\infty} J_j[zf(t-nT_R)] (\exp\{i(j+1)[\omega_l(t-nT_R) + n\phi]\} \\
 & + \exp\{i^2(j-1)[\omega_l(t-nT_R) + n\phi]\}) \sum_{k=-\infty}^{\infty} \cdots \sum_{m=-\infty}^{\infty} \\
 = & -\frac{1}{2} \mu_{21} E_0 \exp[-i\omega_0 t] f(t-nT_R) \sum_{j=-\infty}^{\infty} \{J_{j-1}[zf(t-nT_R)] \\
 & + J_{j+1}[zf(t-nT_R)]\} \exp\{ij[\omega_l(t-nT_R) + n\phi]\} \\
 & \times \sum_{k=-\infty}^{\infty} \cdots \sum_{m=-\infty}^{\infty}.
 \end{aligned} \tag{A12}$$

Using the recursion relation

$$J_g(x) = \frac{x}{2g} [J_{g-1}(x) + J_{g+1}(x)], \tag{A13}$$

one can obtain from Eq. (A12)

$$\begin{aligned}
 \text{term}(n) = & -\frac{1}{2} \mu_{21} E_0 \exp[-i\omega_0 t] f(t-nT_R) \\
 & \times \sum_{j=-\infty}^{\infty} \frac{2j}{zf(t-nT_R)} J_j[zf(t-nT_R)] \exp\{ij[\omega_l(t-nT_R) + n\phi]\} \sum_{k=-\infty}^{\infty} \cdots \sum_{m=-\infty}^{\infty} \\
 = & -\frac{\mu_{21}}{d} \exp[-i\omega_0 t] \sum_{j=-\infty}^{\infty} j \omega_l J_j[zf(t-nT_R)] \exp\{ij[\omega_l(t-nT_R) + n\phi]\} \sum_{k=-\infty}^{\infty} \cdots \sum_{m=-\infty}^{\infty}.
 \end{aligned} \tag{A14}$$

Hence

$$\begin{aligned}
 H_{12} = & -\frac{\mu_{21}}{d} \exp[-i\omega_0 t] \sum_{k=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} \cdots \sum_{j=-\infty}^{\infty} \overbrace{(k\omega_l + m\omega \cdots + j\omega_l)}^{n+1} \\
 & \times \overbrace{J_k[zf(t)] J_m[zf(t-T_R)] \cdots J_j[zf(t-nT_R)]}^{n+1} \\
 & \times \overbrace{\exp[ik\omega_l t] \exp[im[\omega_l(t-T_R) + \phi]] \cdots \exp\{ij[\omega_l(t-nT_R) + n\phi]\}}^{n+1}.
 \end{aligned} \tag{A15}$$

The extensive numerical simulations indicate that it is multiphoton transitions that are in charge of the population accumulations (not shown in the figure). Imposing the condition of the N -photon resonance $\omega_0 = N\omega_l$, $N=1, 2, 3, \dots$ and making the RWA by neglecting the off-resonant or counter-rotating terms in Eq. (A15), i.e.,

$$\begin{aligned}
 H_{12} = & -\frac{\mu_{21}}{d}(n+1)N\omega_i \overbrace{J_N[zf(t)] \cdots J_N[zf(t-nT_R)]}^{n+1} \\
 & \times \exp[inN\omega_i t] \overbrace{\exp[-iNT_R] \cdots \exp[-inNT_R]}^n \\
 & \times \overbrace{\exp[iN\phi] \cdots \exp[iN\phi]}^n.
 \end{aligned}
 \tag{A16}$$

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