Stimulated hyper-Raman adiabatic passage. I. The basic problem and examples

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We discuss various theoretical issues that arise when one extends the conventional stimulated Raman adiabatic passage, involving a pump pulse preceded by a Stokes pulse, to situations in which the pump interaction involves a two-photon transition. As in the simpler cases, it is possible to obtain complete population transfer between an initial state and a targeted final state, if certain general conditions on the pulses are met. We point out important considerations, associated with dynamic Stark shifts and multiphoton ionization, which make successful population transfer more difficult in the multiphoton extension. We illustrate these problems and requirements by considering specific examples of excitation in metastable helium.
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I. INTRODUCTION

The use of stimulated Raman adiabatic passage (STIRAP) [1] to produce efficient population transfer (into a specified excited state of an atom or molecule) with the use of appropriately timed pulse pairs (pump and Stokes radiation) is now well understood theoretically [2] and has been demonstrated experimentally [1].

In the STIRAP procedure, pump and Stokes pulses in special time ordering couple the initial and final states via an intermediate (and unpopulated) state. In principle, we can apply the STIRAP technique to any pair of pulsed interactions, not just those that produce single-photon electric-dipole interactions. Interesting possibilities exist for extending the technique to allow a multiphoton transition as one (or both) of the pulses, so that the usual Raman interaction is replaced by a hyper-Raman interaction. There are practical incentives to examine such techniques, because many molecules of interest require relatively high-energy pump photons (uv or vuv) to reach the first excited electronic states. (The Stokes pulse typically connects this state to a high vibrational level, so optical wavelengths suffice.) It is difficult to provide pulses with adequate power and coherence properties in the vuv, and so it would be very useful if one could achieve this excitation by means of a two-photon transition.

We will discuss the extension of STIRAP to cases in which both pulses are produced by two-photon transitions, as may be expressed by the notation $(2+2)$ to contrast with the notation $(1+1)$ for conventional STIRAP. We also consider, in more detail, the case where the pump interaction is produced by a two-photon transition but the Stokes link is a one-photon transition, as may be expressed by the notation $(2+1)$. These extensions from Raman to hyper-Raman interaction (the acronym STIHRAP seems appropriate) introduce several changes in the Hamiltonian, all of which may be derived in various ways (cf. [3], sect. 14.9, and [4]). Although at first glance such extensions seem obvious and straightforward, closer study shows important differences from $(1+1)$ STIRAP and interesting difficulties.

As will be noted, the generalizations introduce dynamic Stark shifts which, like the two-photon Rabi frequency, are proportional to intensity. This has an important consequence: whereas in the usual excitation of a two-state system the single-photon resonance condition is independent of pulse intensity, with a two-photon transition the dynamic Stark shifts act to force a detuning from (two-photon) resonance. This causes significant deviations from the usual picture of pulsed excitation.

Here we examine some of the issues associated with extending conventional (single-photon) STIRAP to the $(2+1)$ STIHRAP scheme mentioned above. In a companion paper (referred to as paper II [5]) we analyze the sensitivity of the population transfer process to the presence of detunings, both static and dynamic. There we show that the inevitable presence of dynamic Stark shifts in the hyper-Raman STIHRAP implies that, unlike conventional STIRAP, the best population transfer occurs when the laser frequencies are *not* tuned so that the overall two-step process is resonant.

II. THE BASIC STIRAP HAMILTONIAN

To place the more general problem in context, we first review some basic aspects of the simpler conventional STIRAP, with some slight revisions of nomenclature and notation to facilitate the extension.

The basic STIRAP procedure involves three states, labeled 1, 2, and 3, linked by two successive interactions traditionally labeled P (for pump) and S (for Stokes). When the pulses are appropriately timed (Stokes before pump) and satisfy simple constraints (for adiabatic evolution) they can produce complete population transfer from the initial state 1 to the target final state 3.

The simplest implementations are described by a rotating-wave approximation (RWA) Hamiltonian (cf. [3], sect. 14.2). In this approximation each pulse is associated with the interaction Hamiltonian between only one pair of states, as

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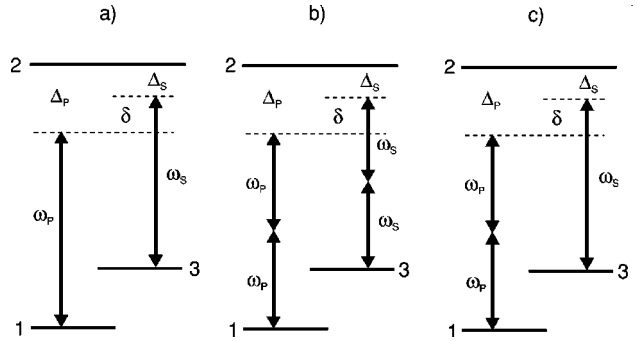

FIG. 1. Diagram of linkages showing definitions of detunings Δ_S, Δ_P, and δ. (a) $(1+1)$ STIRAP, (b) $(2+1)$ STIHRAP, and (c) $(2+2)$ STIHRAP.

symbolized by Fig. 1. There we see that the pump pulse connects states 1 and 2, while the (earlier) Stokes pulse connects states 2 and 3. As is usual, we assume that the energy of state 2 lies above that of the other states 1 and 3 (the so-called Λ configuration), whose relative ranking does not matter. Upon defining a pair of detunings for the carrier frequencies ω_P and ω_S from their assigned Bohr transition frequencies,

$$\hbar\Delta_P \equiv (E_2 - E_1) - \hbar\omega_P, \quad \hbar\Delta_S \equiv (E_2 - E_3) - \hbar\omega_S, \quad (1)$$

and a two-step detuning (this is here just a two-photon detuning, but we generalize subsequently),

$$\delta \equiv \Delta_P - \Delta_S, \quad (2)$$

we can write the basic three-state RWA Hamiltonian matrix as

$$\mathsf{H}(t) = \frac{\hbar}{2}\begin{bmatrix} -\delta & \Omega_P(t) & 0 \\ \Omega_P(t) & \Delta_P + \Delta_S & \Omega_S(t) \\ 0 & \Omega_S(t) & \delta \end{bmatrix}. \quad (3)$$

Although for conventional STIRAP the parameters Δ_S and Δ_P are single-photon detunings and δ is a two-photon detuning, we anticipate more general multiphoton interactions by referring to these as one-step and two-step detunings. For the usual STIRAP, the Rabi frequencies Ω_P and Ω_S are products of dipole moments and electric field amplitudes, such as

$$\begin{aligned} \hbar\Omega_P(t) &= -d_{12}\mathcal{E}_P(t), \\ \hbar\Omega_S(t) &= -d_{32}\mathcal{E}_S(t), \end{aligned} \quad (4)$$

where

$$\begin{aligned} |\mathcal{E}_P(t)|^2 &= (2/c\epsilon_0) I_P(t), \\ |\mathcal{E}_S(t)|^2 &= (2/c\epsilon_0) I_S(t). \end{aligned}$$

The phases and energy zero-point (i.e., the diagonal elements of the RWA Hamiltonian) have here been chosen for subsequent convenience in identifying eigenvalues at early and late times; alternative choices give, as diagonal elements of the RWA Hamiltonian,

$$\begin{aligned} H_{11} &= E_1 + \hbar\omega_P, \\ H_{22} &= E_2, \\ H_{33} &= E_3 + \hbar\omega_S, \end{aligned} \quad (5)$$

or

$$\begin{aligned} H_{11} &= 0, \\ H_{22} &= \hbar\Delta_P, \\ H_{33} &= \hbar\delta. \end{aligned}$$

These forms differ by the inclusion of various overall phase factors common to all quantum states and the choice of zero-point energy.

Of paramount concern here is the creation of conditions which will produce complete population transfer, by suitably arranging pulse intensities, shapes, durations, overlaps, and carrier frequencies. These experimentally controllable properties of the radiation appear parametrized as Rabi frequencies and detunings.

III. THE HYPER-RAMAN HAMILTONIAN

We first consider the $(2+2)$ multiphoton extension of STIRAP in which each pulsed interaction takes place via a two-photon transition. We also consider the simpler extension in which only one multiphoton interaction takes place. We take this to be the pump pulse (generally the excitation energy $E_2 - E_1$ supplied by the pump laser is larger than the energy of the Stokes transition). These generalizations are shown in Fig. 1.

A. The detuning

The detuning for an n-photon transition is the difference between a Bohr frequency and n photon-energy increments $\hbar\omega$. Because we consider a two-photon transition, the definition of the pump detuning becomes

$$\hbar\Delta_P \equiv (E_2 - E_1) - 2\hbar\omega_P. \quad (6)$$

For an n-photon transition, the equation would read $\hbar\Delta_P \equiv (E_2 - E_1) - n\hbar\omega_P$. (A cautionary note: it is common to refer to transitions as one-photon or two-photon. However, the presence of Rabi oscillations or coherent pulses of large area requires coherence among many absorption and emission events. Thus even two-state excitation with one-photon coupling, when coherent, is a multiphoton process.) In the same way, the Stokes detuning for a two-photon transition reads

$$\hbar\Delta_S \equiv (E_2 - E_3) - 2\hbar\omega_S. \quad (7)$$

B. The induced dipole: The polarizability interaction

Whereas for conventional stimulated Raman transitions the two interactions (pump and Stokes) are both obtained from the electric-dipole interaction $V^{E1}(t) = -\mathbf{d}\cdot\mathbf{E}(t)$, the hyper-Raman generalization may be regarded as the interaction between an electric field and an *induced* dipole moment.

The proportionality between the induced moment and the inducing electric field is the (frequency-dependent) polarizability tensor $\alpha(t)$. The resulting interaction takes the form (cf. [3], sect. 14.9) $V^{\rm pol}(t) = -(1/2)\mathbf{E}(t)\cdot\alpha(\omega)\cdot\mathbf{E}(t)$. When placed within a rotating-wave approximation Hamiltonian matrix, diagonal elements of this interaction produce (time-dependent) shifts of energies, while off-diagonal elements give generalizations of the usual Rabi frequencies.

To evaluate the elements of the hyper-Raman interaction, one needs values for the frequency-dependent polarizability tensor. In Cartesian coordinates (appropriate for describing linearly polarized light) the matrix elements of this tensor are evaluated using the formula

$$\langle p'|\alpha_{ij}(\omega)|p\rangle = \sum_q \left[\frac{\langle p'|d_i|q\rangle\langle q|d_j|p\rangle}{(E_q - E_p - \hbar\omega)} + \frac{\langle p'|d_j|q\rangle\langle q|d_i|p\rangle}{(E_q - E_{p'} + \hbar\omega)} \right], \tag{8}$$

where E_p denotes the (unperturbed) energy of an atom in state p. The polarizability involves a pair of dipole transition moments divided by a detuning, and summed over all possible intermediate states. As shown, the sum includes both resonant and antiresonant terms. Although we do not explicitly indicate this, the sums should include a principal-value integral over continuum states. The components d_i of the dipole moment are those selected by the polarization direction of the electric field.

C. The Rabi frequency

In place of the simple product of dipole and field amplitudes that characterizes the two interactions of basic STIRAP, the two-photon Rabi frequency requires the product of a polarizability matrix element and a pair of field amplitudes [i.e., the intensity $I_P(t)$]. For radiation linearly polarized (in the z direction), which we assume, the formula reads

$$\hbar\tilde{\Omega}_P(t) = -\frac{1}{4}\langle 1|\alpha_{zz}(\omega_P)|2\rangle|\mathcal{E}_P(t)|^2 = -\frac{1}{2c\epsilon_0}\langle 1|\alpha_{zz}(\omega_P)|2\rangle I_P(t), \tag{9a}$$

$$\hbar\tilde{\Omega}_S(t) = -\frac{1}{4}\langle 2|\alpha_{zz}(\omega_S)|3\rangle|\mathcal{E}_S(t)|^2 = -\frac{1}{2c\epsilon_0}\langle 2|\alpha_{zz}(\omega_S)|3\rangle I_S(t). \tag{9b}$$

It is generally important for the success of STIRAP that the two peak Rabi frequencies be roughly equal. One can perhaps balance a weak single-photon transition (e.g., a ''forbidden'' transition) with the two-photon transition, or one can use a weaker laser for the one-photon transition. Of course, the condition of large pulse area must still apply.

D. The dynamic Stark shifts

Real atoms or molecules are never exactly only three-level structures. The remaining energy levels, in the presence of radiation, act to shift the three energy levels which comprise the STIHRAP system. Like the two-photon Rabi frequency, these shifts are (to a first approximation) proportional to the product of an atomic polarizability and a field intensity. When computing shifts it is important to consider the effect of *each* field upon *each* transition, i.e., both the Stokes laser and the pump laser cause shifts of both the initial and final level. We denote the shift in energy of state i caused by pulse a as $\hbar S_{ia}$. This shift can be computed from appropriate components $\alpha_{ij}(\omega)$ of the frequency-dependent polarizability tensor and the intensity,

$$\hbar S_{ia}(t) = -\frac{1}{2c\epsilon_0}\langle i|\alpha_{zz}(\omega_a)|i\rangle I_a(t). \tag{10}$$

It is important to note that dynamic Stark shifts are proportional to intensity. In conventional STIRAP the Rabi frequencies are proportional to the square root of intensity, and therefore it is possible to adjust intensity such that Rabi frequencies are large (mainly because of near-resonant coupling) but Stark shifts are small. With a two-photon transition, the Rabi frequencies and the Stark shifts scale together with intensity; it is not possible to eliminate the shifts by adjusting the intensity.

Usually the final state lies higher in energy than the initial state, and it has larger polarizability and the larger Stark shifts. However, this is not always the case. The presence of a nearby energy level or the occurrence of very different dipole moments may cause the initial state to have the larger shifts.

Dynamic Stark shifts have been included (and proven essential) when the coupling of the initial and final states is via a continuum [6]. To the best of our knowledge, their consequences have not been considered when the coupling is between bound states.

E. The (2+2) Hamiltonian

With the inclusion of dynamic Stark shifts and two-photon Rabi frequencies, the (2+2) RWA Hamiltonian takes the form

$$\mathsf{H}(t) = \frac{\hbar}{2}\begin{bmatrix} -\delta + 2[S_{1P}(t) + S_{1S}(t)] & \tilde{\Omega}_P(t) & 0 \\ \tilde{\Omega}_P(t) & \Delta_P + \Delta_S + 2[S_{2P}(t) + S_{2S}(t)] & \tilde{\Omega}_S(t) \\ 0 & \tilde{\Omega}_S(t) & \delta + 2[S_{3P}(t) + S_{3S}(t)] \end{bmatrix}. \tag{11}$$

Because the shifts, like the transition-interaction operator, involve polarizabilities, a rigorous derivation of this Hamiltonian (e.g., using adiabatic multimode Floquet theory) [3,4] will provide consistent expressions for all elements of the Hamiltonian.

We can simplify the appearance of the Hamiltonian by writing it in terms of Stark-shifted single-step detunings,

$$\tilde{\Delta}_P(t)=\Delta_P+[S_{2S}(t)+S_{2P}(t)]-[S_{1S}(t)+S_{1P}(t)], \tag{12}$$

$$\tilde{\Delta}_S(t)=\Delta_S+[S_{2S}(t)+S_{2P}(t)]-[S_{3S}(t)+S_{3P}(t)], \tag{13}$$

and a shifted two-step detuning

$$\tilde{\delta}(t)=\Delta_P-\Delta_S+[S_{3S}(t)+S_{3P}(t)]-[S_{1S}(t)+S_{1P}(t)]. \tag{14}$$

When so written, the $(2+2)$ Hamiltonian reads

$$\mathsf{H}(t)=\frac{\hbar}{2}\begin{bmatrix} -\tilde{\delta}(t) & \tilde{\Omega}_P(t) & 0 \\ \tilde{\Omega}_P(t) & \tilde{\Delta}_P(t)+\tilde{\Delta}_S(t) & \tilde{\Omega}_S(t) \\ 0 & \tilde{\Omega}_S(t) & \tilde{\delta}(t) \end{bmatrix}. \tag{15}$$

This matrix has the same formal expression as the basic matrix (3) for three-state STIRAP.

As can be seen from the present choice of diagonal elements, the excitation behavior of the atom is determined by the differences of Stark shifts. Note that both the pump and the Stokes pulses contribute to each of the single-step detunings, even when, as in the RWA, each pulse is in (near) resonance with only one of the transitions. Note also that for the trapped state to be an eigenstate of this Hamiltonian, it is necessary that the Stark-shifted two-step detuning $\tilde{\delta}$ should vanish throughout the interaction, which is usually not the case because the two interactions have different time dependences.

The STIRAP process does not require that the intermediate state be resonant. Judging from the experience with conventional $(1+1)$ STIRAP, one might think that Stark shifts of this state are not important, and that it would be essential that the two-step process of pump and Stokes be resonant (as expressed by the condition $\delta=0$). This is not quite correct. Satisfactory population transfer can occur so long as the two-step detuning $\tilde{\delta}$ remains within appropriate limits and this depends on the detuning of the pump laser from the shifted resonance. Shifts which increase this detuning will make transfer more difficult. In paper II [5] we examine this sensitivity to detuning.

F. The (2+1) Hamiltonian

A potential application of hyper-Raman STIHRAP occurs when the pump transition requires a large energy change, and therefore takes place via a two-photon transition, but the Stokes interaction remains the conventional one-photon transition [see Fig. 1(b)]. The resulting $(2+1)$ hyper-Raman Hamiltonian

$$\mathsf{H}(t)=\frac{\hbar}{2}\begin{bmatrix} -\tilde{\delta}(t) & \tilde{\Omega}_P(t) & 0 \\ \tilde{\Omega}_P(t) & \tilde{\Delta}_P(t)+\tilde{\Delta}_S(t) & \Omega_S(t) \\ 0 & \Omega_S(t) & \tilde{\delta}(t) \end{bmatrix} \tag{16}$$

is a variant of Eq. (15), but with $\Omega_S(t)$ in place of $\tilde{\Omega}_S(t)$ and with dynamic detunings defined as

$$\tilde{\Delta}_P(t)=\Delta_P+S_{2P}(t)-S_{1P}(t), \tag{17}$$

$$\tilde{\Delta}_S(t)=\Delta_S+S_{2P}(t)-S_{3P}(t), \tag{18}$$

with $\hbar\Delta_S=E_2-E_3-\hbar\omega_S$ and $\hbar\Delta_P=E_2-E_1-2\hbar\omega_P$. Thus only the pump field contributes to the dynamic Stark shifts.

G. Spontaneous emission losses

In real atoms all energy levels except the ground state have some probability (however small) for spontaneous radiative decay. This spontaneous emission represents a loss of probability from the decaying state. It has two other effects: it appears as a growth of population in some state (possibly the initial or final state of the STIHRAP process), and it causes diminution (relaxation) of coherences between states. It is usually the case that, during the STIRAP process, there occurs negligible decay of either the initial state (1) or the final target state (3). When such decays are present, they limit the population which can be placed into the target state. However, often the lifetime of the intermediate state (2) is sufficiently short that its decay must be considered. If this decay does not go appreciably to states 1 or 3, then the decay can be modeled with a complex-valued energy, leading to an imaginary contribution to the (2,2) element of the $(2+2)$ Hamiltonian (15), which becomes

$$\mathsf{H}(t)=\frac{\hbar}{2}\begin{bmatrix} -\tilde{\delta}(t) & \tilde{\Omega}_P(t) & 0 \\ \tilde{\Omega}_P(t) & \tilde{\Delta}_P(t)+\tilde{\Delta}_S(t)-i\gamma_2 & \tilde{\Omega}_S(t) \\ 0 & \tilde{\Omega}_S(t) & \tilde{\delta}(t) \end{bmatrix}. \tag{19}$$

The $(2+1)$ Hamiltonian differs only in the replacement of $\tilde{\Omega}_S(t)$ with $\Omega_S(t)$.

Although the strictly resonant ($\delta=0$) excitation case has a trapped state for any value of pump detuning and intermediate-state loss γ_2, in practice one must consider a range of two-step detunings for which population transfer occurs despite the lack of coincidence between the state vector and an exact trapped state. The possibility of loss from the intermediate state has a significant effect upon this linewidth, which is proportional to $\Omega/\sqrt{\gamma_2\tau}$ [7]. The adiabatic condition $|\Omega\tau|\gg 1$ must be supplemented with the condition

$$(\Omega\tau)^2\gg\gamma_2\tau. \tag{20}$$

H. Induced losses: Photoionization

When the energy of a pulse photon ($\hbar\omega_P$ or $\hbar\omega_S$) exceeds the binding energy of any of the three STIRAP states, then the pulse can produce photoionization. This acts as a

population loss, and can be modeled as an (additional) imaginary term on the diagonal of the Hamiltonian matrix.

Even though a single photon may not have enough energy to produce ionization, a two-photon ionization may occur. This interaction, like the interaction which produces the two-photon Rabi frequency and the dynamic Stark shifts, is the product of an atomic polarizability and a field intensity. Let the loss produced from state i by pulse a be Γ_{ia}. For n-photon photoionization this loss can be parametrized by a generalized cross section $\sigma_i^{(n)}$ in the form

$$\Gamma_{ia}=(I_a)^n\sigma_i^{(n)}. \tag{21}$$

When such laser-induced losses are included, the $(2+2)$ Hamiltonian takes the form

$$\mathsf{H}=\frac{\hbar}{2}\begin{bmatrix} -\tilde{\delta}-i(\Gamma_{1P}+\Gamma_{1S}) & \tilde{\Omega}_P & 0 \\ \tilde{\Omega}_P & \tilde{\Delta}_P+\tilde{\Delta}_S-i(\gamma_2+\Gamma_{2P}+\Gamma_{2S}) & \tilde{\Omega}_S \\ 0 & \tilde{\Omega}_S & \tilde{\delta}-i(\Gamma_{3P}+i\Gamma_{3S}) \end{bmatrix}. \tag{22}$$

The $(2+1)$ Hamiltonian differs only in the replacement of $\tilde{\Omega}_S(t)$ with $\Omega_S(t)$, possibly also dropping terms Γ_{nS}.

Loss from the intermediate state is not detrimental so long as exact two-step resonance is maintained. However, the detrimental effect of intermediate-state loss noted above for nonresonant cases will be even more pronounced when losses due to photoionization are included.

One must be sure that the pump pulse, which remains active after population transfer has occurred, does not photoionize the final state, either by a one-photon transition or by a two-photon transition. One must also be sure that there are no states which the pump laser can connect, near resonantly, to the final state. If there is such a connection, one must consider not just the three-state STIRAP, but a four-state system. The final-state interaction may cause not only loss but also, by Autler-Townes splitting, may prevent the needed STIRAP resonance.

It is also important that the Stokes laser does not ionize the initial state, either by one-photon or by two-photon excitation. Such loss will deplete the population which is available for STIRAP. One must also be sure that there is no additional level for which the Stokes pulse will be near resonance with the initial state.

I. Additional final states

It may happen that the target state is one of several closely spaced states. For example, when there is a single nearby energy level, coupled by electric-dipole radiation to the intermediate state, the $(2+2)$ Hamiltonian takes the form

$$\mathsf{H}=\frac{\hbar}{2}\begin{bmatrix} -\tilde{\delta}-i(\Gamma_{1P}+\Gamma_{1S}) & \tilde{\Omega}_P & 0 & 0 \\ \tilde{\Omega}_P & \tilde{\Delta}_P+\tilde{\Delta}_S-i(\gamma_2+\Gamma_{2P}+\Gamma_{2S}) & \tilde{\Omega}_S & \tilde{\Omega}_S' \\ 0 & \tilde{\Omega}_S & \tilde{\delta}-i(\Gamma_{3P}+i\Gamma_{3S}) & 0 \\ 0 & \tilde{\Omega}_S' & 0 & \tilde{\delta}_4-i(\Gamma_{4P}+i\Gamma_{4S}) \end{bmatrix}. \tag{23}$$

The difference between δ and δ_4 is the separation between the two neighboring levels, each competing for the interaction of the Stokes pulse with the intermediate state (2). The STIRAP mechanism can be initiated if the target is sufficiently separated from this nearest neighbor. The separation must exceed the two-step bandwidth.

J. Pulse pairs

Here we carry out modeling of the $(2+1)$ STIHRAP using Gaussian pulses. We use the amplitude

$$f(t)=\exp[-(t/\tau)^2]. \tag{24}$$

This pulse is normalized to have area $\int dt f(t)=\sqrt{\pi}\tau$; the full width at half maximum of $f(t)^2$ is 1.825τ. We take the pump and Stokes Rabi frequencies to be

$$\Omega_S(t)=\Omega_{\max}f(t-\tau_S/2),\quad \tilde{\Omega}_P(t)=\Omega_{\max}f(t+\tau_S/2)^2 \tag{25}$$

thereby defining the time delay τ_S of the Stokes pulse with respect to the pump pulse (τ_S is negative for a counterintuitive sequence). Figure 2 sketches this sequence. Because the pump interaction is proportional to the square of the electric field amplitude, the associated (two-photon) pump Rabi fre-

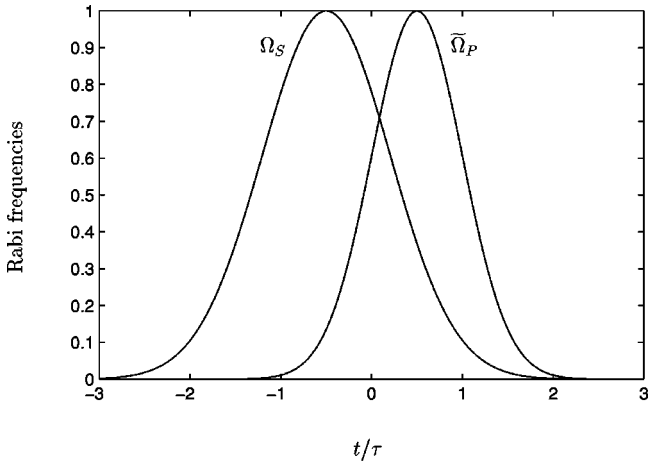

FIG. 2. Amplitudes of Rabi frequencies vs time. First pulse is $\Omega_S(t)$, second pulse is $\widetilde{\Omega}_P(t)$.

quency has a more sharply peaked pulse shape than is the case for the pump Rabi frequency of ordinary STIRAP (see Fig. 2).

IV. EXAMPLES

The preceding section presented the Hamiltonian needed to describe hyper-Raman STIRAP (or STIHRAP). We illustrate the typical concerns by presenting two examples, both of excitation of helium. The first example, for which we present numerical simulations, offers a good opportunity for achieving population transfer.

The second example illustrates some of the detrimental effects which can make experimental implementation of STIHRAP unsuccessful.

A. A successful example in helium

As a first example, we examine a possible $(2+1)$ STIHRAP in metastable helium, based on linearly polarized light connecting $M=0$ sublevels. Starting from the initial state $2s\ ^3S_1$, one proceeds via a two-photon transition to the $3s\ ^3S_1$ state, and then on to the final target state $2p\ ^3P_0$ or $2p\ ^3P_2$, in the sequence

$$2s\ ^3S_1 \leftrightarrow 3s\ ^3S_1 \leftrightarrow 2p\ ^3P_{0,2}.$$

Figure 3 shows the energy levels of this system.

We have used a simple model potential [8] as the basis for obtaining an atomic wave function from which to evaluate the various polarizability matrix elements. In Table I, computed values for relevant Stark shifts and Rabi frequencies are given.

Unlike conventional STIRAP, where the choice $\delta=0$ is always preferable, the presence of dynamic Stark shifts makes an alternative choice of detuning preferable. Figure 4 shows an example of the time evolution of population in the three-level system with $2p\ ^3P_0$ as the target level, for appropriately chosen detunings and pulse delays. The losses from level 2 out of the three-level system include spontaneous decay to the levels $2p\ ^3P_1$ and $2p\ ^3P_2$ (giving $\Gamma = 0.0246\ \mathrm{ns}^{-1}$) and two-photon ionization by the pump laser

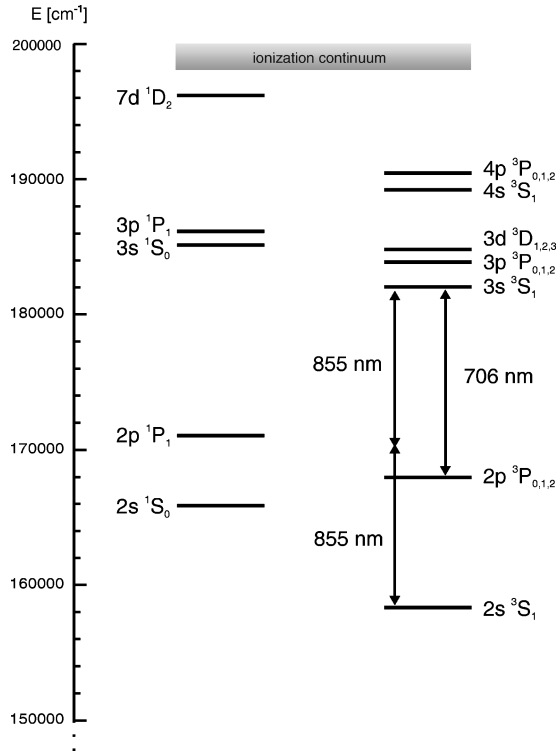

FIG. 3. Energy levels of helium showing hyper-Raman pump transition via two 855 nm photons and Stokes transition via 706 nm photon.

($\gamma = 0.6\times 10^{-10}\times I_p^2\ \mathrm{s}^{-1}$). As can be seen, population transfer is successful, even in the presence of dynamic Stark shifts.

Figure 5 shows how the population transfer efficiency $P_3(\infty)$ depends on pulse delay. The characteristic feature of a STIRAP process is clearly visible: the Stokes pulse must precede the pump pulse for good transfer efficiency.

In paper II [5], we examine this system in more detail, giving a theoretical foundation for the particular choice of parameters (pulse delay and detunings) chosen. We also present an analysis of the sensitivity of the population transfer to the choice of one- and two-step detunings.

Experiments to confirm this analysis are underway.

B. An unsuccessful example in helium

To illustrate some of the difficulties which may prevent $(2+1)$ STIHRAP, we present an example of another excitation of metastable helium. Starting from the initial state $2s\ ^1S_0$, one proceeds via two-photon transition to the $7d\ ^1D_2$ state, and then on to the final target state $2p\ ^3P_1$, in the sequence

TABLE I. Parameters for helium. Intensity is expressed in W/cm^2 and shifts in s^{-1}.

State	Shift	Rabi frequency
$1s2s\ ^3S_1$	$141\ I_P$	
$1s3s\ ^3S_1$	$118\ I_P$	$\widetilde{\Omega}_P = 152\ I_P$
$1s2p\ ^3P_0$	$-143\ I_P$	$\Omega_S = 1.61\times 10^8\sqrt{I_S}$
$1s2p\ ^3P_2$	$-220\ I_P$	$\Omega_S = 2.28\times 10^8\sqrt{I_S}$

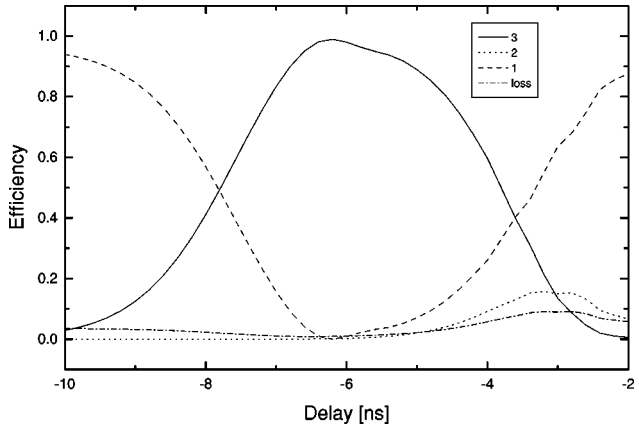

FIG. 4. Population histories $P_n(t)$ for $(2+1)$ hyper-Raman STIHRAP in helium. The pulse intensities are $I_P=250\ \mathrm{MW/cm^2}$ and $I_S=0.0557\ \mathrm{MW/cm^2}$ giving the peak Rabi frequency $\Omega_{\max}=38\ \mathrm{ns^{-1}}$. The pulse width (for pump and Stokes) is 5 ns (full width at half maximum of laser intensity). Delay is $\tau_S=-6.0$ ns. Spontaneous emission loss from state 2 is $\Gamma=0.0246\ \mathrm{ns^{-1}}$. Two-photon ionization by the pump laser is $\gamma=0.003\,75\ \mathrm{ns^{-1}}$. Static detunings are $\delta=37\ \mathrm{ns^{-1}}$ and $\Delta_S+\Delta_P=11\ \mathrm{ns^{-1}}$.

$$2s\ ^1S_0 \leftrightarrow 7ds\ ^1D_2 \leftrightarrow 2p\ ^3P_1.$$

Although at first thought the scheme appears to be a good choice, closer study reveals serious problems.

The Stokes coupling of the intermediate $7d\ ^1D$ level and the target $2p\ ^3P$ is a weak one to better balance the coupling strength for the two-photon pump transition. However, the Stokes photons act also within the triplet manifold and couple the intended final state to the $7d\ ^3D$ levels. This coupling strength exceeds the (intermediate to final) coupling (^1D-^3P) by orders of magnitude. Consequently, whatever population reaches $2p\ ^3P$ will be strongly coupled to $7d\ ^3D$. This is not at all a three-state system. In addition, the strong Stark splitting may alter the mixing with the $7d\ ^1D$ state and may thus change the $7d\ ^1D-2p\ ^3P$ coupling strength.

Furthermore, the pump photons use the $3s\ ^3S$ state as a stepping stone for efficient ionization. At intensities needed to saturate the $2s\ ^1S-7d\ ^1D$ transition, the pump laser ionizes the final ($2p\ ^3P$) state very efficiently. Thus, even if all of what was said above were discounted, one would find little population in the final state: either the coupling at the first step is too weak or—if some transfer takes place—it readily ionizes. The ionization of the $7d\ ^1D$ state by the pump photon may also be detrimental.

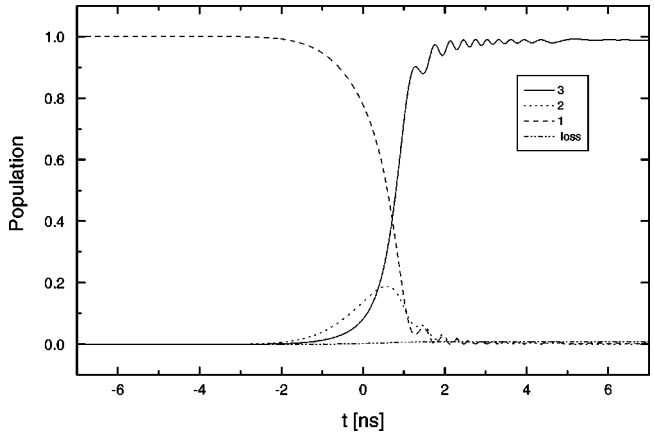

FIG. 5. Dependence of transfer efficiency $P_3(\infty)$ upon pulse delay for $(2+1)$ hyper-Raman STIHRAP in helium. Parameters are as in Fig. 4.

V. CONCLUSIONS

We have considered coherent population transfer by stimulated hyper-Raman adiabatic passage (STIHRAP) for the case when the coupling between the initial and intermediate states is by a two-photon transition. Although the use of a two-photon transition in place of a one-photon transition introduces few novel mathematical distinctions, the physics of coherently driven population transfer can be quite different. Because the Rabi frequency is, like the dynamic Stark shifts, proportional to intensity and polarizability matrix elements, it is essential to consider the effects of such shifts. They can dramatically alter the scenario for successful population transfer and are often detrimental. We have shown two examples, one potentially successful and the other illustrating various detrimental effects which will prevent the successful completion of population transfer.

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