Detrimental consequences of small rapid laser fluctuations on stimulated Raman adiabatic passage

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We discuss the detrimental effect of small rapid random fluctuations of laser-field amplitude and phase upon the efficiency of stimulated Raman adiabatic passage (STIRAP). Such fluctuations typically accompany the laser stabilization procedures that produce nearly monochromatic light on top of a much broader bandwidth Lorentz-profile pedestal which may carry only a few percent or less of the total power. As we will show, their effects differ qualitatively from the fluctuations that have hitherto been considered (for example, phase diffusion). We present analytic expressions for the population transfer efficiency of STIRAP when limited by stochastic fluctuations of this type. These expressions show, in contrast to situations discussed in the past in which population transfer improves with increasing peak Rabi frequencies, that for the weak broadband noise that accompanies a strong narrow-spectral component, there is an optimum value for the peak Rabi frequency and that the effect of fluctuations, although small, cannot be entirely eliminated in practice. The mission of the current work is to point out that, under the given circumstances, efforts in experiments trying to overcome the detrimental consequences of fluctuations by increasing the intensity, which is the intuitively proper approach, will not be successful.

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I. INTRODUCTION

A. STIRAP

Stimulated Raman adiabatic passage (STIRAP) has been intensively investigated for more than 20 years (see reviews in [1–5] and references therein) as a technique for transferring population adiabatically between two selected quantum states. As a view of current literature shows, the STIRAP procedure has proven very useful for an increasing variety of purposes in physics, chemistry, and engineering: more than a hundred articles on this subject appear each year. In its most basic implementation, the STIRAP technique exposes a three-state quantum system to two sequential partially overlapping transform-limited laser pulses. Initially, the population resides in state $|1\rangle$, and the intended target of the population, state $|3\rangle$, is entirely unpopulated. An excited state $|2\rangle$ serves as an intermediary for the population transfer, although it acquires only a negligible population at all times. The first pulse to appear, S, or Stokes, has a constant carrier frequency ω_S close to the transition frequency ω_{23} between states $|3\rangle$ and $|2\rangle$ while the constant carrier frequency ω_P of the later pulse, P, or pump, is close to the transition frequency ω_{12} of the transition between states $|1\rangle$ and $|2\rangle$. The two carrier frequencies must satisfy the two-photon resonance condition between initial and final states,

$$\omega_P - \omega_S = \omega_{12} - \omega_{23}, \tag{1}$$

where ω_{nm} is the transition frequency between states n and m.

STIRAP has proven particularly useful for producing ultracold molecules by photoassociation (see, e.g., [6–12]). Experiments aiming at the formation of cold molecules in their lowest quantum state (see, e.g., [13–20]) have, at times, encountered unexpected difficulties in achieving anticipated high efficiencies, despite experimental conditions that meet traditional requirements for laser power and bandwidth.

Whereas transfer efficiencies of 90%–95% are often considered satisfactory for selective quantum-state preparation in atoms and molecules, more recent interest in quantum information processing [21,22] using STIRAP [23–29] calls for the errors of gate operations to be 10^{-4} or less [22,30]. It is thus important to understand possible limitations that would prevent such achievements by STIRAP.

In this paper we examine one source of unanticipated difficulty in achieving high efficiency with STIRAP: a noise source that cannot be overcome by common remedies for inefficiencies, such as increasing the laser power. Figure 1 shows a spectral line shape typical of those observed for stabilized diode lasers: a narrow central component atop a much smaller broad pedestal (cf. [31]). This line shape is the basis of our subsequent analysis.

B. Adiabatic requirements and limitations

Successful population transfer with STIRAP requires that the state vector adiabatically follows the dark, or population trapping, adiabatic state but does not require specific pulse shapes or intensities, apart from general constraints imposed to ensure adiabatic evolution: the process is insensitive to small variations in pulse amplitudes, widths, delay, and single-photon detuning. This robustness of the STIRAP process has prompted studies that guide the design of optimized pulses that, given experimental limitations on pulse duration and intensity, produce the highest transfer efficiency [30,32–41].

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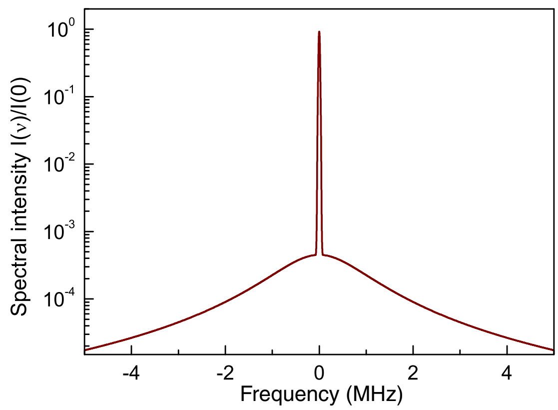

FIG. 1. (Color online) Spectral line shape, typical for stabilized diode lasers, assumed in the present analysis.

There has been much work, both theoretical and experimental, elucidating the conditions needed for complete population transfer within the STIRAP procedure (see reviews [1–3,5]). A condition for adiabatic following, for transform-limited pulses, is that for each pulse the product of peak Rabi frequency and pulse duration (the temporal pulse area) should be much larger than π and that population transfer becomes increasingly complete as the pulse areas become larger. The most favorable situation occurs when the two pulses each fulfill a single-photon resonance condition, $\omega_P = \omega_{12}$ and $\omega_S = \omega_{23}$, but these conditions are not necessary: What matters is the two-photon resonance condition Eq. (1). The consequences of violating these conditions were analyzed in [42,43]. It is also desirable for the peak Rabi frequencies to be the same for both fields (within 10%–20%). In the analysis below we will assume that the two peak values are equal. However, our basic conclusions do not depend on this assumption.

Early work showed that if only three nondegenerate states are involved and if the pulse envelopes are smooth and the pulse areas are sufficiently large for each pulse, then the population transfer can be very nearly complete. The detrimental effects of near degeneracies [44], additional levels [45–49], and a high density of molecular energy levels [50] have been discussed.

In the present paper we extend earlier work, discussed in the following sections, that treated the effects of various noise sources on the STIRAP process. We show that even with well-stabilized laser fields there can remain a limit on the success of the STIRAP population transfer.

C. Fluctuations

Irregular uncontrollable changes of the environment of a quantum system, i.e., fluctuations and noise, affect the success of any designed quantum procedure such as STIRAP. These fluctuations enter the rotating-wave approximation (RWA) time-dependent Schrödinger equation through detunings and Rabi frequencies. Although the Rabi frequency derives entirely from the excitation field and therefore incorporates such fluctuations as it may have, the detuning derives from not only the laser frequency but also the transition frequencies, and these undergo fluctuations that originate with the environment, including variations in neighboring atoms. Such environmental effects are often treated by means of density-matrix equations in which there occur relaxation terms that affect, at different rates, the populations (diagonal elements of the density matrix) and the coherences (off-diagonal elements). Here we do not follow that approach but deal directly with a model of the fluctuating field amplitude and frequency.

The STIRAP procedure relies on the creation and maintenance of a dark state of the system, a superposition of initial and final states $|1\rangle$ and $|3\rangle$, with the composition set by the two fields, that has no linkage to the lossy excited state $|2\rangle$. The effects of typical laser-field fluctuations on this dark state were considered by Dalton and Knight well before the development of STIRAP [51,52]. Using a δ-correlated phase-diffusion model of the laser field (a Wiener-Levy, or W-L, stochastic process [53], associated with a Lorentz spectral profile), they found that laser fluctuations dephased the atomic coherences, with consequent loss of the population trapping associated with the dark state. They saw that fluctuations removed the narrow coherence minimum in the absorption spectrum at the two-photon resonance. However, they also found that when the driving fields are critically cross-correlated in their fluctuations (e.g., by deriving one field from the other so that any laser jitter in the interaction zone is common to both beams), then the two-photon coherences are unaffected by laser fluctuations and the coherence minimum persists. The favorable effects of cross-correlation on STIRAP were reported, e.g., in Refs. [54–57].

It is important to realize that although it is convenient to characterize the noise properties of a laser by a single number, the laser bandwidth, this simplification may be inadequate for predicting effects produced by the laser: a variety of phase and amplitude variations can produce the same bandwidth, yet have very different effects upon excitation, fluorescence, and other laser-induced processes [53,58–60].

In the early papers on STIRAP there was already concern about the effect of laser fluctuations and how these would affect the need to maintain adiabatic evolution in order to transfer population successfully. Kuhn *et al.* [61] carried out the first numerical simulation of STIRAP with a noisy laser. They used a stochastic model of zero-mean exponentially correlated noise (an Ornstein-Uhlenbeck, or O-U, process [53]). They assumed no cross-correlation of the two fields. Because the O-U process incorporates two parameters (the spectral density of the noise D and the correlation time of fluctuations), the laser bandwidth alone is not sufficient to define the process fully. Predictions based on the O-U process can be more realistic, but they require more complete characterization of the laser noise than just bandwidth.

The Ornstein-Uhlenbeck model of the laser fluctuations was used in an extensive examination of the STIRAP process by Yatsenko and coworkers [43,54–56]. They found that with this type of laser fluctuation it was always possible, in principle, to improve population transfer for a fixed pulse duration by increasing the peak Rabi frequencies.

Among the other recent papers that have discussed the detrimental effect of stochastic processes on STIRAP we note Refs. [62–73].

D. Present work

Stabilizing the lasers is a natural procedure for reducing fluctuations and diminishing nonadiabatic losses, but our analysis in the present work shows that even with such refinements the transfer efficiency may fall well below the desired value of unity.

The present work examines how a small stochastic modulation of the almost-monochromatic laser fields affects the STIRAP efficiency. Such a field is emitted, for example, by stabilized diode lasers [31]. When the narrowing of a laser spectrum is accomplished with the technique of stabilization to a high-finesse cavity, the resulting laser radiation spectrum consists of a very narrow line (with a width typically much less than 1 MHz, seemingly ideal for STIRAP) on a wide background pedestal (with a width of 1 MHz or more), as shown in Fig. 1. Typically, the total power of the wide background is less than 10% of the total laser power. Hitherto, the consequence of the noise related to the line shape shown in Fig. 1 on the population transfer efficiency of STIRAP has, to the best of our knowledge, not been considered.

We here adopt the means of describing laser fluctuations used earlier [54], but we use a different model for the underlying stochastic process, one that models the nature of fluctuations that remain even when the laser has been carefully stabilized. In contrast to earlier work, we will here show that, when the field has a small broadband stochastic component, increasing the peak Rabi frequency with a fixed pulse duration may not improve the STIRAP efficiency. Although the effects of nonadiabaticity diminish with increasing peak Rabi frequency, the stochastic contributions saturate. Consequently, as we will show, there is an optimum peak Rabi frequency.

II. BASIC EQUATIONS

We consider a three-state quantum system with excitation linkages in the usual Λ form: two long-lived, low-energy states, states $|1\rangle$ and $|3\rangle$, each linked via electric dipole interaction to an excited state $|2\rangle$.

We assume that the spontaneous radiative decay of the excited state, which occurs at rate γ, goes entirely to states other than $|1\rangle$ and $|3\rangle$ and that there are no interactions directly linking $|1\rangle$ and $|3\rangle$. The quantum system interacts, via dipole transition moments $\mathbf{d}$, with the electric field $\mathbf{E}(t)$ of two pulses, termed Stokes (S) and pump (P):

$$\mathbf{E}(t) = \tfrac{1}{2}[\mathbf{E}_P(t)\exp(-i\omega_P t - i\varphi_P) + \mathbf{E}_S(t)\exp(-i\omega_S t - i\varphi_S) + \text{c.c.}]. \quad (2)$$

These have phases φ_P and φ_S and carrier frequencies ω_P and ω_S that are close to the $1 \leftrightarrow 2$ and $2 \leftrightarrow 3$ transition frequencies, respectively.

We model the pulse amplitudes $\mathbf{E}_P(t)$ and $\mathbf{E}_S(t)$ as superpositions of strong, smooth, nonfluctuating real-valued functions of time $\mathbf{E}_{P0}(t)$ and $\mathbf{E}_{S0}(t)$ and small stochastic complex-valued additive fields $\varepsilon_P(t)\mathbf{E}_{P0}(t)$ and $\varepsilon_S(t)\mathbf{E}_{S0}(t)$:

$$\mathbf{E}_j(t) = [1+\varepsilon_j(t)]\mathbf{E}_{j0}(t), \qquad j = P,S. \quad (3)$$

With this model the phases φ_P and φ_S are constant; without loss of generality, we set these equal to zero.

We assume the statistical behavior of $\varepsilon_j(t) = \varepsilon'_j(t) + i\varepsilon''_j(t)$ to be

$$\begin{aligned} \langle\varepsilon_j(t)\rangle &= 0,\\ \langle\varepsilon'_i(t)\varepsilon''_j(t')\rangle &= 0,\\ \langle\varepsilon'_i(t)\varepsilon'_j(t')\rangle &= \varepsilon_{j0}^{(ampl)2}\exp\left(-G_j^{(ampl)}|t-t'|\right)\delta_{i,j},\\ \langle\varepsilon''_i(t)\varepsilon''_j(t')\rangle &= \varepsilon_{j0}^{(phase)2}\exp\left(-G_j^{(phase)}|t-t'|\right)\delta_{i,j},\\ i,j &= P,S, \end{aligned} \quad (4)$$

where angle brackets denote ensemble averages and $\delta_{i,j}$ is the Kronecker delta. We introduced here the superscripts $(ampl)$ and $(phase)$ to stress that for weak modulation the real part $\varepsilon'_j(t)$ is the stochastic amplitude modulation index and the imaginary part $\varepsilon''_j(t)$ can be considered to be a small phase modulation:

$$[1+\varepsilon_j(t)]\mathbf{E}_{j0} \simeq [1+\varepsilon'_j(t)]\exp[i\varepsilon''_j(t)]\mathbf{E}_{j0}. \quad (5)$$

Note that this phase modulation is different from the phase fluctuations described by the O-U process. Due to the statistical behavior (4) the total phase is not diffusing as it is for the O-U process, but instead, it fluctuates around zero.

The optical spectrum of the field (3), when the amplitudes are constant, consists of a narrow central component that is almost monochromatic (treated as a δ function) and two broad backgrounds of Lorentzian shape with the widths $G_j^{(ampl)}$ and $G_j^{(phase)}$. These carry the fraction

$$\varepsilon_0^2 = \varepsilon_{j0}^{(phase)2} + \varepsilon_{j0}^{(ampl)2} \ll 1 \quad (6)$$

of the total laser power. In real lasers the widths $G_j^{(ampl)}$ and $G_j^{(phase)}$ are almost always equal. Hereafter, for the sake of simplicity, we will assume

$$G_j^{(ampl)} = G_j^{(phase)} = G_J, \quad \varepsilon_{0j}^{(phase)2} = \varepsilon_{0j}^{(ampl)2} = \varepsilon_{j0}^2/2. \quad (7)$$

Figure 1 shows an example of this spectrum with the width of the background $G_j = 2\pi \times 1$ MHz and with the total noise power equal to 2% of the total laser power ($\varepsilon_{j0}^2 = 0.02$).

As is customary, we adopt the RWA. The three components of the state vector form a three-component column vector $\mathbf{C}(t)$ with elements $\{C_1(t),C_2(t),C_3(t)\}$ that obeys the time-dependent Schrödinger equation,

$$\frac{d}{dt}\mathbf{C}(t) = -i\mathsf{W}(t)\mathbf{C}(t), \quad (8)$$

where $\mathsf{W}(t)$ is a 3×3 RWA Hamiltonian matrix. For our assumed one- and two-photon resonance this reads

$$\mathsf{W}(t) = \frac{1}{2}\begin{bmatrix} 2\dot{\varepsilon}''_P(t) & \Omega_P(t)[1+\varepsilon'_P(t)] & 0\\ \Omega_P(t)[1+\varepsilon'_P(t)] & -i\gamma & \Omega_S(t)[1+\varepsilon'_S(t)]\\ 0 & \Omega_S(t)[1+\varepsilon'_S(t)] & 2\dot{\varepsilon}''_S(t) \end{bmatrix}. \quad (9)$$

Here the off-diagonal elements, the Rabi frequencies,

$$\Omega_P(t) = -\langle 1|\mathbf{d}\cdot\mathbf{E}_{P0}(t)|2\rangle/\hbar,$$
$$\Omega_S(t) = -\langle 3|\mathbf{d}\cdot\mathbf{E}_{S0}(t)|2\rangle/\hbar, \tag{10}$$

are slowly varying real-valued functions of time.

It is convenient to express the RWA Hamiltonian matrix in an alternative basis, using the time-dependent "bright" state $\Phi_b(t)$ and "dark" state $\Phi_d(t)$,

$$\begin{bmatrix}\Phi_b(t)\\ \Phi_2(t)\\ \Phi_d(t)\end{bmatrix} = \begin{bmatrix}\sin\theta(t) & 0 & \cos\theta(t)\\ 0 & 1 & 0\\ \cos\theta(t) & 0 & -\sin\theta(t)\end{bmatrix}\begin{bmatrix}|1\rangle\\ |2\rangle\\ |3\rangle\end{bmatrix}. \tag{11}$$

The time-dependent mixing angle $\theta(t)$, defined through the relationship

$$\tan\theta(t) = \frac{\Omega_P(t)[1+\varepsilon'_P(t)]}{\Omega_S(t)[1+\varepsilon'_S(t)]}, \tag{12}$$

involves the noisy modulations term, and as a result, it has a smooth part and a noisy part. With our assumed pulse sequence of Stokes before pump, the dark state $\Phi_d(t)$ coincides initially with the initially populated bare state $|1\rangle$, and it aligns after the pulse sequence with the target state $|3\rangle$. Thus by maintaining the state vector $\Psi(t)$ in this dark state at all times we accomplish the population transfer $|1\rangle \to |3\rangle$ of a traditional STIRAP process.

The expansion of the state vector in this basis reads

$$\Psi(t) = C_b(t)\Phi_b(t) + C_2(t)\Phi_2(t) + C_d(t)\Phi_d(t). \tag{13}$$

After some algebra we obtain the time-dependent Schrödinger equation for the column vector $\mathbf{C}^{(BD)}(t)$ with components $\{C_b(t), C_2(t), C_d(t)\}$,

$$i\frac{d}{dt}\mathbf{C}^{(BD)}(t) = \mathsf{W}^{(BD)}(t)\mathbf{C}^{(BD)}(t), \tag{14}$$

where the RWA Hamiltonian matrix in the bright-dark basis reads (with suppression of explicit notation of time dependence)

$$\mathsf{W}^{(BD)}\mathbf{C}^{(BD)} = \frac{1}{2}\begin{bmatrix} 0 & \Omega_{\rm rms} & 2i\dot\theta + (\dot\varepsilon''_P - \dot\varepsilon''_S)\sin 2\theta\\ \Omega_{\rm rms} & -i\gamma & 0\\ -2i\dot\theta + (\dot\varepsilon''_P - \dot\varepsilon''_S)\sin 2\theta & 0 & 0\end{bmatrix}\begin{bmatrix}C_b\\ C_2\\ C_d\end{bmatrix}. \tag{15}$$

Here the rms Rabi frequency $\Omega_{\rm rms}(t)$ is

$$\Omega_{\rm rms}(t) = \sqrt{\Omega_P(t)^2[1+\varepsilon'_P(t)]^2 + \Omega_S(t)^2[1+\varepsilon'_S(t)]^2}. \tag{16}$$

III. ANALYTICAL RESULTS

A. Formulation

The equations for the state amplitudes read

$$\frac{d}{dt}C_d = \left[-\dot\theta - i\tfrac{1}{2}(\dot\varepsilon''_P - \dot\varepsilon''_S)\sin 2\theta\right]C_b,$$
$$\frac{d}{dt}C_b = \left[\dot\theta - i\tfrac{1}{2}(\dot\varepsilon''_P - \dot\varepsilon''_S)\sin 2\theta\right]C_d - i\tfrac{1}{2}\Omega_{\rm rms}C_2,$$
$$\frac{d}{dt}C_2 = -i\tfrac{1}{2}\Omega_{\rm rms}C_b - \tfrac{1}{2}\gamma C_2. \tag{17}$$

Two processes will produce changes in the dark-state amplitude: the nonadiabatic coupling term $\dot\theta$ that is present with any pulses and an explicitly stochastic term.

Due to the coupling between the dark state and the bright state the population of the dark state decreases during the STIRAP process. We assume the dark-state losses to be small, and we take $C_d = 1$ in the equations for the amplitude C_b and C_2. The resulting equations read

$$\frac{d}{dt}C_b = -i\tfrac{1}{2}\Omega_{\rm rms}C_2 + \dot\theta - i\tfrac{1}{2}(\dot\varepsilon''_P - \dot\varepsilon''_S)\sin 2\theta,$$
$$\frac{d}{dt}C_2 = -\tfrac{1}{2}\gamma C_2 - i\tfrac{1}{2}\Omega_{\rm rms}C_b. \tag{18}$$

The presence of decay in Eq. (15) allows one to find solutions to the Schrödinger equation using perturbation theory. This was done for nonfluctuating fields in Ref. [74] for the two-photon resonant case and in Ref. [43] for nonresonant case. We will again use that approach.

B. Solution procedure

Equations (18) describe a damped harmonic oscillator driven by the stochastic force. They can be solved for arbitrary $\theta(t)$, $\Omega_{\rm rms}(t)$, and $\varepsilon(t)$. The result is

$$C_b(t) = i\int_{-\infty}^{t} dt'\,\frac{\Omega_{\rm rms}(t')\left\{\dot\theta(t') - \frac{i}{2}[\dot\varepsilon''_P(t') - \dot\varepsilon''_S(t')]\sin 2\theta(t')\right\}}{\Omega(t')\Omega_{\rm rms}(t)}[\lambda^*(t)F(t,t') - \lambda(t)F^*(t,t')], \tag{19}$$

$$C_2(t) = -\int_{-\infty}^{t} dt'\,\frac{\Omega_{\rm rms}(t')\left\{\dot\theta(t') - \frac{i}{2}[\dot\varepsilon''_P(t') - \dot\varepsilon''_S(t')]\sin 2\theta(t')\right\}}{2\Omega(t')}[F(t,t') - F^*(t,t')], \tag{20}$$

where

$$\Omega(t) = \sqrt{\Omega_{\rm rms}(t)^2 - \frac{1}{4}\gamma^2}, \quad \lambda(t) = -\frac{1}{4}\gamma + \frac{i}{2}\Omega(t),$$
$$F(t,t') = \exp\left(\int_{t'}^{t} \lambda(t'')\,dt''\right). \tag{21}$$

Substituting $C_b(t)$ from Eq. (19) into the first equation of (17), we find the dark-state derivative $\dot{C}_d(t)$. We integrate this to obtain the dark-state amplitude $C_d(t)$. The probability of successful population transfer via STIRAP is the final value of dark-state population, averaged over fluctuations

$$P_{\rm STIRAP} = \langle |C_d(\infty)|^2 \rangle. \tag{22}$$

This probability differs from unity for two reasons: there is a (small) probability P_A that the evolution is not adiabatic, and there is a (small) probability P_N that the broadband noise prevents success. It is convenient to evaluate the small noise-dependent difference P_N between $P_{\rm STIRAP}$ and what would be obtained with completely adiabatic evolution by writing

$$P_{\rm STIRAP} = 1 - P_N - P_A \equiv 1 - P_{\rm loss}. \tag{23}$$

To evaluate the noise contribution P_N we assume complete adiabaticity. Then the required expression is

$$P_N \approx 1 - \langle |C_d(\infty)|^2 \rangle = 2\,{\rm Re} \int_{-\infty}^{\infty} \left\langle \left\{ \dot{\theta}(t) + \frac{i}{2}[\dot{\varepsilon}''_P(t) - \dot{\varepsilon}''_S(t)]\sin 2\theta(t) \right\} C_b(t) \right\rangle dt$$
$$= i \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \left\langle \frac{2\Omega_{\rm rms}(t')\left\{\dot{\theta}(t) + \frac{i}{2}[\dot{\varepsilon}''_P(t) - \dot{\varepsilon}''_S(t)]\sin 2\theta(t)\right\}\left\{\dot{\theta}(t') - \frac{i}{2}[\dot{\varepsilon}''_P(t') - \dot{\varepsilon}''_S(t')]\sin 2\theta(t')\right\}}{\Omega(t')\Omega_{\rm rms}(t)} \right\rangle$$
$$\times [\lambda^*(t)F(t,t') - \lambda(t)F^*(t,t')]. \tag{24}$$

We are here examining the almost ideal STIRAP process in which there are negligible nonadiabatic losses in the absence of any noise. The functions $F(t,t')$ assure that the time difference $t - t'$ is much less than the variation time of Rabi frequencies, meaning that we can take $\Omega_{\rm rms}(t') = \Omega_{\rm rms}(t)$ and $\Omega(t') = \Omega(t)$. Finally, we neglect $\dot{\theta}_0(t)$, defined as the smooth part of the mixing angle,

$$\tan\theta_0(t) = \Omega_P(t)/\Omega_S(t). \tag{25}$$

Then we obtain

$$P_N = \int_{-\infty}^{\infty} dt \frac{2i}{\Omega(t)} \int_{-\infty}^{t} dt' \left\langle \left\{ \delta\dot{\theta}(t) + \frac{i}{2}[\dot{\varepsilon}''_P(t) - \dot{\varepsilon}''_S(t)]\sin 2\theta_0(t) \right\} \left\{ \delta\dot{\theta}(t') - \frac{i}{2}[\dot{\varepsilon}''_P(t') - \dot{\varepsilon}''_S(t')]\sin 2\theta(t') \right\} \right\rangle$$
$$\times [\lambda^*(t)F(t,t') - \lambda(t)F^*(t,t')]. \tag{26}$$

We assume small modulation $|\varepsilon_{S,P}| \ll 1$ so that

$$\delta\dot{\theta} = \dot{\theta} - \dot{\theta}_0 \simeq \sin\theta_0\cos\theta_0[\dot{\varepsilon}'_P - \dot{\varepsilon}'_S]. \tag{27}$$

With this assumption we obtain, for the noise contribution to the loss of STIRAP success, the expression

$$P_N = \int_{-\infty}^{\infty} dt\, \frac{2i\sin^2\theta_0\cos^2\theta_0}{\Omega(t)} \int_{-\infty}^{t} dt'\, [\langle \dot{\varepsilon}_P(t)\dot{\varepsilon}^*_P(t')\rangle + \langle \dot{\varepsilon}_S(t)\dot{\varepsilon}^*_S(t')\rangle][\lambda^*(t)F(t,t') - \lambda(t)F^*(t,t')]. \tag{28}$$

Using the correlation behavior of Eq. (4), we obtain finally the basic formula for the effect of small broadband noise on the STIRAP success:

$$P_N = 2\int_{-\infty}^{+\infty} dt\; \sin^2\theta_0(t)\cos^2\theta_0(t)\Omega^2_{\rm rms}(t)\left[\frac{G_S}{4G_S^2 + 2G_S\gamma + \Omega_{\rm rms}(t)^2}\varepsilon^2_{S0} + \frac{G_P}{4G_P^2 + 2G_P\gamma + \Omega_{\rm rms}(t)^2}\varepsilon^2_{P0}\right]. \tag{29}$$

This expression depends upon the specific pulses only through the variation of the rms Rabi frequency and the variation of the traditional mixing angle. It sums separate contributions from the two fields. It can be evaluated numerically for any pulse shape $\Omega_{\rm rms}(t)$.

C. Saturation

It is instructive to examine the limit when, during an interval from $-T$ to T, the rms Rabi frequency is very large. The two fractions then simplify, and one has the approximation

$$P'_N \approx 2\left[G_S\varepsilon^2_{S0} + G_P\varepsilon^2_{P0}\right]\int_{-T}^{+T} dt\; \sin^2\theta_0(t)\cos^2\theta_0(t). \tag{30}$$

This is independent of the peak Rabi frequency: increasing this parameter will not improve the STIRAP transfer.

D. Model results

To obtain an analytical expression we took (as mentioned above) the S and P amplitudes to be of equal magnitude and shape, offset in time by t_d,

$$\Omega_S(t) = \Omega_0 f(t), \qquad \Omega_P(t) = \Omega_0 f(t - t_d), \tag{31}$$

and we used the following specific expression for the pulse-shape function $f(t)$:

$$f(t) = \begin{cases} \cos[\frac{\pi}{2}\frac{t}{\tau}] & -\tau < t < \tau, \\ 0 & \text{otherwise.} \end{cases} \tag{32}$$

For the pulse shape (32) with $t_d = \tau$ the rms Rabi frequency is a constant, $\Omega_{\rm rms} = \Omega_0$, during the time interval when both pulses act. Then the result (29), expressed as a function of the peak Rabi frequency Ω_0, reads

$$P_N(\Omega_0) = \frac{1}{4}\Omega_0^2\left[\frac{G_S\tau}{4G_S^2 + 2G_S\gamma + \Omega_0^2}\varepsilon_{S0}^2 + \frac{G_P\tau}{4G_P^2 + 2G_P\gamma + \Omega_0^2}\varepsilon_{P0}^2\right]. \tag{33}$$

For very large peak Rabi frequency this probability saturates and cannot be smaller than

$$P_N(\Omega_0) \to \tfrac{1}{4}\left[G_S\tau\varepsilon_{S0}^2 + G_P\tau\varepsilon_{P0}^2\right]. \tag{34}$$

E. Phase fluctuation noise

For comparison, the losses in the case of the two free-running independent lasers with the phase fluctuations considered in [54] are expressible as

$$P_N^{(phase)} = \int_{-\infty}^{+\infty} dt\, \sin^2[2\theta(t)]\left[\frac{D_P^{(ph)}G_P^{(ph)}\left(2G_P^{(ph)}+\gamma\right)}{\Omega_{\rm rms}(t)^2+4G_P^{(ph)2}+2\gamma G_P^{(ph)}} + \frac{D_S^{(ph)}G_S^{(ph)}\left(2G_S^{(ph)}+\gamma\right)}{\Omega_{\rm rms}(t)^2+4G_S^{(ph)2}+2\gamma G_S^{(ph)}}\right], \tag{35}$$

where $D_j^{(ph)}$ and $G_J^{(ph)}$ are the parameters of the Ornstein-Uhlenbeck process describing the phase fluctuations. For the pulse shape (32) the integral in (35) can be evaluated analytically. The result is

$$P_N^{(phase)}(\Omega_0) = \frac{1}{4}\frac{D_P^{(ph)}G_P^{(ph)}\left(2G_P^{(ph)}+\gamma\right)\tau}{\Omega_0^2+4G_P^{(ph)2}+2\gamma G_P^{(ph)}} + \frac{1}{4}\frac{D_S^{(ph)}G_S^{(ph)}\left(2G_S^{(ph)}+\gamma\right)\tau}{\Omega_0^2+4G_S^{(ph)2}+2\gamma G_S^{(ph)}}. \tag{36}$$

This probability loss becomes arbitrarily small as the peak Rabi frequency increases.

$$P_N^{(phase)}(\Omega_0) \to \frac{1}{4\Omega_0^2}\left[D_P^{(ph)}G_P^{(ph)}\left(2G_P^{(ph)}+\gamma\right)\tau + D_S^{(ph)}G_S^{(ph)}\left(2G_S^{(ph)}+\gamma\right)\tau\right]. \tag{37}$$

F. Nonadiabatic losses

The detrimental effects of nonadiabatic evolution, as distinct from noise, are parametrized by the probability P_A. These can be written as [54]

$$P_A(\Omega_0) = \int_{-\infty}^{+\infty} 4\gamma\frac{\dot\theta^2}{\Omega_{\rm rms}(t)^2}dt\,. \tag{38}$$

For the pulse shape (32) the integral (38) can be evaluated analytically. The result is

$$P_A(\Omega_0) = \frac{\gamma\pi^2}{\Omega_0^2\tau}\,. \tag{39}$$

This loss diminishes monotonically with increasing peak Rabi frequency, and therefore, in principle, it can be made arbitrarily small.

IV. DISCUSSION AND CONCLUSION

Coherence losses caused by the different mechanisms discussed here depend on the maximum Rabi frequency Ω_0 and interaction time τ in different ways. The nonadiabaticity losses decrease as Ω_0 and τ increase. Note that for a short-lived exited state, for which $\gamma\tau \gg 1$, the losses are not exponentially small as one expects for adiabatic evolution of a long-lived state. Nevertheless, by using laser pulses which are strong and sufficiently long one can reach noise-free STIRAP efficiency close to 100%.

A similar situation occurs if the laser phase fluctuations are an O-U process. In this case one can improve the STIRAP efficiency by increasing the Rabi frequency but not the interaction time. Such laser fluctuations could be neglected for stabilized lasers when $D_j^{(ph)} \ll \gamma,\Omega_0$.

Unfortunately, one cannot decrease the contribution of the fast small fluctuations described by the effective modulation index ε_0 by increasing the Rabi frequency Ω_0. Figure 2 shows an example of the dependence of the nonadiabatic and noise modulation losses on the Rabi frequency Ω_0 calculated for typical experimental conditions.

Our results show that there is an optimum Rabi frequency for use with STIRAP: the efficiency decreases for either

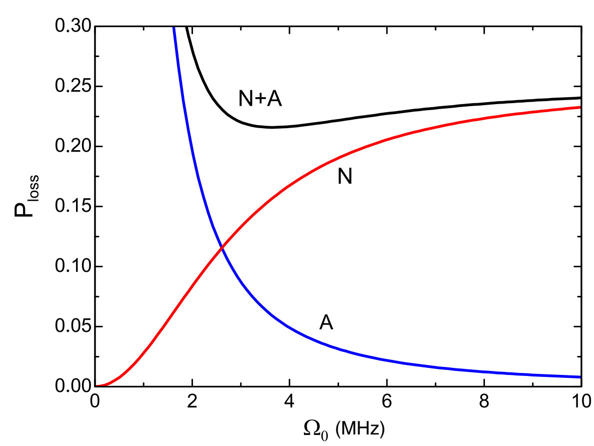

FIG. 2. (Color online) The STIRAP efficiency loss $P_{\rm loss}(\Omega_0)$ for the pulse shape (32) as a function of the maximum Rabi frequency Ω_0. Curve N is the contribution $P_N(\Omega_0)$ from broadband noise, Eq. (33), and curve A is the contribution $P_A(\Omega_0)$ from nonadiabaticity, Eq. (39). Parameters are $\gamma = 2\pi \times 2$ MHz, $G_S = G_P = 2\pi \times 1$ MHz, $\tau = 4$ μs, $\varepsilon_{S0}^2 = \varepsilon_{P0}^2 = 0.02$.

larger or smaller values. There is no other way to overcome the problem of weak fluctuations of phase and amplitude fluctuations, except by eliminating the spectral pedestal: one cannot overcome the detrimental consequences of the fluctuations by using larger Rabi frequencies. The mission of the current work is to point out that, under the given circumstances, efforts in experiments trying to overcome the detrimental consequences of fluctuations by increasing the intensity, which is the intuitively proper approach, will not be successful

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