Atom manipulation based on delayed laser pulses in three- and four-level systems: Light shifts and transfer efficiencies

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Atomic mirrors and beam splitters based on adiabatic following by delayed laser pulses in a three-level Raman configuration are analyzed. We show that in a pure three-level system of two ground states and one excited state, laser pulses tuned precisely on resonance with the excited state do not produce any accumulated phase shifts due to ac Stark shifts even if the process is nonadiabatic. A numerical simulation suggests that multiple delayed laser pulses are attractive for precision interferometry since high transfer efficiencies (up to 98.7% per pulse for the cesium $D1$ line) and low ac Stark shifts are expected. A possible application to optical two-photon spectroscopy for an optical clock is discussed.

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I. INTRODUCTION

Laser light has been used to coherently split and recombine atomic wave packets in atomic interferometer experiments [1]. A particularly useful method is light pulse interferometry based on stimulated Raman transitions connecting sharp hyperfine ground states [2]. The sensitivity of such a measurement may be increased by applying multiple π pulses ("atomic mirrors"), introducing an additional recoil of $2\hbar k$ per pulse to the interferometer. In a recent measurement of the cesium recoil energy, the photon recoil has been increased by sandwiching up to 15 additional Raman π pulses between the two sets of $\pi/2$ pulses, giving rise to a recoil splitting of 64 photon momenta between two interferometers of opposite recoil shift [3,4]. The observed transfer efficiency per pulse was limited to about 85% by spatial variations in the beam intensity, so that perfect π pulses can not be generated for all the atoms. Atoms left behind in the wrong atomic state after each π pulse did not affect the fringe contrast since they could be distinguished and removed from the interferometer before the final beam splitter. On the other hand, a large number of imperfect π pulses reduces the signal size.

Population transfer by coherent adiabatic passage is not very sensitive to variations in the beam intensities and should allow higher transfer efficiencies. Also, the obtained transfer is expected to be very robust for a realistic laser power stability. In this paper, we investigate a method of population transfer based on stimulated Raman adiabatic passage (STIRAP) [5–8]. This method has been demonstrated first by Gaubatz *et al.* [6,7], and its application to atomic interferometry has been proposed by Marte, Zoller, and Hall [8]. Recently, population and momentum transfer by delayed laser pulses has been observed in atomic systems [9,10].

Adiabatic transfer based on changes of light intensities for precision experiments have the advantage over adiabatic transfers relying on a sweep of the (two-photon) resonance frequency [11,12], that the two-photon resonance condition may be maintained all the time for the STIRAP method. In this scheme, the transfer of population between ground states $|1\rangle$ and $|3\rangle$ via an excited state $|2\rangle$ can be done adiabatically with time delayed laser pulses of light at frequency ω_1 tuned to the $|1\rangle \rightarrow |2\rangle$ transition and ω_2 tuned to the $|3\rangle \rightarrow |2\rangle$ transition, as shown in Fig. 1. For given Rabi frequencies Ω_1 at ω_1 and Ω_2 at ω_2, there is a superposition state that is "dark," i.e., not connected to the excited state $|2\rangle$. For example, for $\Omega_1=0$ the dark state is $|1\rangle$, and for $\Omega_1=\Omega_2$ the dark state contains states $|1\rangle$ and $|3\rangle$ with equal amplitudes. An atom initially in $|1\rangle$ can be transferred adiabatically to $|3\rangle$ by first turning on Ω_2 with $\Omega_1=0$ and then increasing Ω_1 as shown in Fig. 2. We later discuss possible atomic beam splitters based on this method and show the setup of a proposed interferometer using adiabatic following.

Adiabatic following by delayed laser pulses appears attractive since the atoms remain in a dark state and should not undergo any ac Stark shifts. However, transfer in a finite amount of time requires slight nonadiabaticity and the atoms undergo many Rabi cycles during the transfer. We discuss here adiabatic transfer for real atomic systems, paying particular attention to the ac Stark shifts induced during the transfer process. We show analytically that in a pure three-level system, the atoms do not experience any ac Stark shift when the lasers are tuned precisely on resonance with the intermediate state. This condition is also not particularly sensitive to slight errors in detuning.

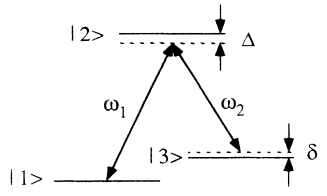

FIG. 1. Level scheme for population transfer by adiabatic following between two ground states via an excited state. For counterpropagating laser beams, the momenta of two optical photons per transition is transferred to the atom.

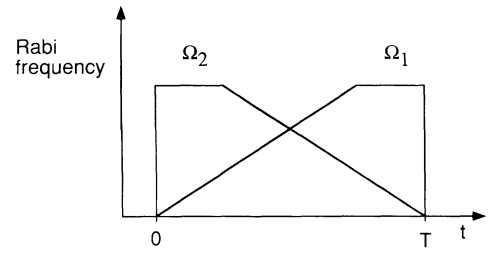

FIG. 2. Pulse sequence for efficient population transfer between two ground states $|1\rangle$ and $|3\rangle$ via an excited state $|2\rangle$ by adiabatic following, where light of frequency ω_1 couples levels $|1\rangle$, $|2\rangle$ (Rabi frequency Ω_1) and ω_2 couples $|2\rangle$, $|3\rangle$ (Rabi frequency Ω_2). For transfer from $|1\rangle$ to $|3\rangle$, the dark state is $|1\rangle$ if $\Omega_1(t=0)=0$. At some intermediate time when $\Omega_2=\Omega_1$, the dark state consists of equal amplitudes of $|1\rangle$ and $|3\rangle$, and when $\Omega_2(t=T)=0$ the system is left completely in state $|3\rangle$.

In real atoms, the presence of additional excited off-resonant states will introduce some ac Stark shift and limit the maximum possible transfer efficiency. In a recent experiment using the cesium $D2$ line, about 40% of the atoms were coherently transferred from $6S_{1/2}(F=4)$, $m_F=4$ to $m_F=-4$ [10]. In this work it was suggested that the imperfect transfer was due to off-resonant excitation in the excited state hyperfine manifold. Here, we here consider the cesium $D1$ line, which has the advantage of a 5.8 times larger excited state hyperfine splitting (1.17 GHz for $6P_{1/2}$) [13]. Off-resonant excitation thus is expected to be suppressed by a factor of about $(5.8)^2=34$ compared to the $D2$ line. Our numerical simulations also show that the relative matrix elements between the different hyperfine states are much more favorable in the case of the $D1$ line. Further, we have considered transitions between magnetic field insensitive states that are necessary for precision interferometer systems [2].

By numerical integration of the Schrödinger equation, we find that this adiabatic transfer method is superior to off-resonant Raman transitions in terms of transfer efficiency and ac Stark shifts.

II. GENERAL FORMULAS

We consider a three-level atom with a level structure as shown in Fig. 1 irradiated by two counterpropagating laser beams of optical frequencies ω_1 and ω_2 connecting the states $|1\rangle\leftrightarrow|2\rangle$ and $|2\rangle\leftrightarrow|3\rangle$, respectively. The Hamiltonian for the system is

$$H_{\text{total}}=\frac{p_{\text{op}}^2}{2m}+\hbar\left[\omega_{21}^A-\frac{i\Gamma}{2}\right]|2\rangle\langle 2| +\hbar\omega_{31}^A|3\rangle\langle 3|-e\mathbf{r}\cdot\mathbf{E}\,, \tag{1}$$

where $\hbar\omega_{21}^A$ and $\hbar\omega_{31}^A$ denote the atomic energy spacings between level $|1\rangle$ and levels $|2\rangle$ and $|3\rangle$, respectively. The relaxation of the intermediate state $|2\rangle$ has been accounted for by introducing a non-Hermitian term $-i\Gamma/2$ as in Ref. [8]. We neglect decay into levels $|1\rangle$ and $|3\rangle$ by spontaneous emission since it causes only an incoherent background to the fringes. This assumption is verified by numerically solving the Bloch equations.

The electric field of the two incident beams is assumed to be

$$\mathbf{E}=\mathbf{E}_{1,0}\cos(k_1z-\omega_1t+\phi_1) +\mathbf{E}_{2,0}\cos(-k_2z-\omega_2t+\phi_2)\,. \tag{2}$$

For counterpropagating ($k\sim k_1\sim k_2$) laser beams $\sim 2\hbar k$ momentum per transition is transferred to the atoms. We assume that the laser detunings from the respective transition frequencies are small compared to the splittings between the states ($\omega_1\sim\omega_{21}^A$ and $\omega_2\sim\omega_{21}^A-\omega_{31}^A$) so that transitions are induced only in a closed family of momentum states. The time evolution of its wave function

$$|\Psi\rangle=c_1(p,t)\exp\left[-i\frac{p^2}{2m\hbar}t\right]|1\rangle+c_2(p+\hbar k_1,t)\exp\left[-i\left[\frac{(p+\hbar k_1)^2}{2m\hbar}+\omega_{21}^A\right]t\right]|2\rangle +c_3(p+\hbar k_1+\hbar k_2,t)\exp\left[-i\left[\frac{(p+\hbar k_1+\hbar k_2)^2}{2m\hbar}+\omega_{31}^A\right]t\right]|3\rangle \tag{3}$$

can be described by the time evolution of the coefficients $c_i(p_i,t)$. The equations of motion for the interaction wave function

$$|\varphi\rangle=\begin{bmatrix} c_1(p,t)\\ c_2(p+\hbar k_1,t)\\ c_3(p+\hbar k_1+\hbar k_2,t)\end{bmatrix} \tag{4}$$

is determined by the Hamiltonian

$$H=-\frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_1 e^{-i(\phi_1+t\Delta)} & 0\\ \Omega_1 e^{i(\phi_1+t\Delta)} & i\Gamma & \Omega_2 e^{i[\phi_2+t(\Delta+\delta)]}\\ 0 & \Omega_2 e^{-i[\phi_2+t(\Delta+\delta)]} & 0\end{bmatrix}, \tag{5}$$

where

$$\Omega_1=\frac{e}{\hbar}\langle 2|r\cdot\mathbf{E}_{1,0}|1\rangle ,$$

$$\Omega_2=\frac{e}{\hbar}\langle 2|r\cdot\mathbf{E}_{2,0}|3\rangle ,$$

$$\Delta=-(\omega_1-\omega_{21}^{A})+\frac{1}{2m\hbar}((p+\hbar k_1)^2-p^2) , \quad (6)$$

$$\delta=(\omega_1-\omega_2-\omega_{31}^{A})+\frac{1}{2m\hbar}(p^2-(p+\hbar k_1+\hbar k_2)^2) ; \quad (7)$$

see Refs. [14,15]. For the case when both detunings are equal to zero, the interaction Hamiltonian simplifies to

$$H=-\frac{\hbar}{2}\begin{pmatrix} 0 & \Omega_1 e^{-i\phi_1} & 0 \\ \Omega_1 e^{i\phi_1} & i\Gamma & \Omega_2 e^{i\phi_2} \\ 0 & \Omega_2 e^{-i\phi_2} & 0 \end{pmatrix} . \quad (8)$$

The effect of small detunings Δ and δ from the one- and two-photon resonances are discussed in Ref. [6]. Generally speaking, the following formulas are still approximately valid as long as both detunings are small compared to Ω_{eff} and unless otherwise noted, we will assume $\delta=\Delta=0$.

When the populations and coherences change adiabatically, steady state solutions can be applied [7]. For the laser pulse shapes as shown in Fig. 2, adiabatic following requires

$$\Omega_{\text{eff}}T\gg 1 , \quad (9)$$

where $\Omega_{\text{eff}}=\sqrt{\Omega_1^2+\Omega_2^2}$ denotes an effective Rabi frequency. Under these conditions, one of the three eigenvectors of the system is

$$|v_1(t)\rangle=\begin{pmatrix} \cos\theta(t) \\ 0 \\ -\sin\theta(t)e^{-i(\phi_2-\phi_1)} \end{pmatrix} , \quad (10)$$

where $\tan\theta(t)=\Omega_1(t)/\Omega_2(t)$. This eigenvector is of special interest, since for this state the amplitudes of both lower levels for absorption into the excited state precisely cancel [16]. This "dark state" is not coupled to the excited level and since the eigenvalue of this state is equal to zero, it does not undergo any ac Stark shift. Adiabatic change of the dark state allows a complete population transfer from $|1\rangle$ to $|3\rangle$.

Our numerical solutions solve the full quantum-mechanical problem and therefore include the slight nonadiabatic coupling to the excited state. The pulse shape shown in Fig. 2 (for $\Omega_{1,\max}=\Omega_{2,\max}$) allows faster transfer with the same efficiency as for the two displaced Gaussian pulses of Ref. [7]. Note that as long as all of the population is in one of the ground states, the laser addressing the transition connecting the other ground state with the excited state may be switched on and off as quickly as possible.

So far, we mainly have considered complete population transfer. The effect of a pulse sequence on an initial wave function $|\varphi(-\infty)\rangle$ acting as an optical element via adiabatic following can, in the limit of very long interaction times T ($\Omega_{\text{eff}}T\gg 1$ and $\Gamma T\gg 1$), be written as

$$|\varphi(T)\rangle=|v_1(T)\rangle\langle v_1(0)|\varphi(-\infty)\rangle . \quad (11)$$

Equation (11) suggests that an adiabatic following beam-splitter can be accomplished by simultaneously turning both lasers into zero intensity in the middle of a pulse sequence as shown in Fig. 2 (after $\Omega_1=\Omega_2$), as noted in Ref. [8]. The opposite time sequence may be used for the final beam splitter in an atomic interferometer, where two atomic wave packets are recombined. This element may also be used for redirecting the wave packets when the two arms of the interferometer are in different states, as necessary for the second pulse of a four $\pi/2$ pulse [17] interferometer. Figure 3 shows the arrangement and (simplified) pulse sequence for an atomic interferometer using adiabatic following in four interaction regions.

A possible fourth "optical element" with two input and two output channels can be formed by simply turning on and off both lasers simultaneously. In the dressed picture, this element is a projector onto the dark state and an optical analog to a Stern-Gerlach apparatus similar to recent work in a two-level system [18] where, however, only one dressed state is transmitted. For an atom initially in one of the ground states, this element generates a coherent superposition of the two ground states, where,

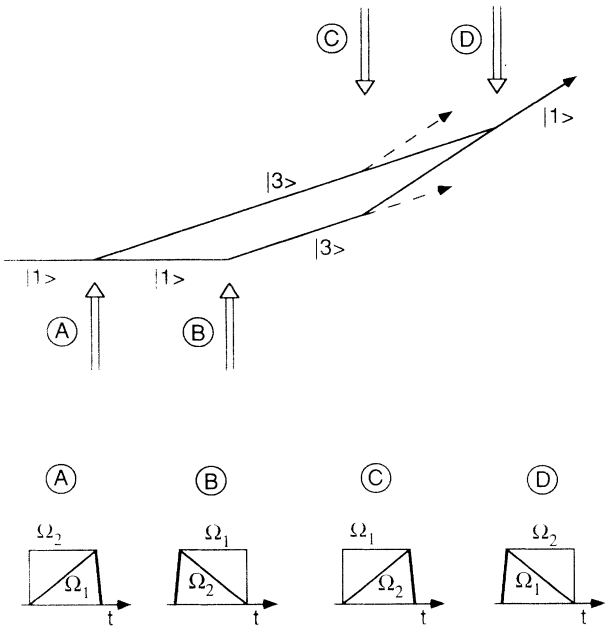

FIG. 3. Pulse sequence for an atomic interferometer based on adiabatic following by delayed laser pulses; for the level scheme see Fig. 1. The double line shows the propagation axis of the light with frequency ω_1 (Rabi frequency Ω_1). Light with frequency ω_2 (Rabi frequency Ω_2) is directed oppositely. The analogous interferometer with opposite recoil shift is obtained by applying pulse sequence *D* instead of *B* in the second interaction region, and pulse sequence *A* instead of *C* in the third interaction region. If atoms in state $|3\rangle$ instead of $|1\rangle$ are desired at the output of the interferometer, pulse sequence *B* should be used in the fourth interaction region.

however, half the population is lost due to spontaneous emission. We have "demonstrated" all four optical elements by numerical simulation in a three-level system.

III. ANALYTIC RESULTS

During the population transfer, the atomic states undergo many Rabi oscillations and the nonadiabatic part may *a priori* allow a nonnegligible phase shift to accumulate. We now show that as long as both Δ and δ are zero there is no accumulated phase shift for arbitrary pulse forms. Assume the initial wave function before the laser pulses is

$$|\varphi(-\infty)\rangle = \begin{bmatrix} d_1(-\infty) \\ id_2(-\infty)e^{i\phi_1} \\ d_3(-\infty)e^{-i(\phi_2-\phi_1)} \end{bmatrix}, \tag{12}$$

with all coefficients $d_i(-\infty)$ real. Then the wave function for any time t can be written as

$$|\varphi(t)\rangle = \begin{bmatrix} d_1(t) \\ id_2(t)e^{i\phi_1} \\ d_3(t)e^{-i(\phi_2-\phi_1)} \end{bmatrix}, \tag{13}$$

with all $d_i(t)$ real.

We can prove (13) by direct substitution into the time-dependent Schrödinger equation, which gives

$$\dot{d}_1(t) = -\tfrac{1}{2}\Omega_1(t)d_2(t) , \tag{14a}$$

$$\dot{d}_2(t) = \tfrac{1}{2}(\Omega_1(t)d_1(t) - \Gamma d_2(t) + \Omega_2(t)d_3(t)) , \tag{14b}$$

$$\dot{d}_3(t) = -\tfrac{1}{2}\Omega_2(t)d_2(t) . \tag{14c}$$

The coupled system of first-order differential equation has the form

$$\dot{\mathbf{d}}(t) = A(t)\mathbf{d}(t) , \tag{15}$$

where $A(t)$ is a real matrix. For all t, we can separate $\mathbf{d}(t)$ into real and imaginary parts as

$$\mathbf{d}(t) = \mathbf{u}(t) + i\mathbf{v}(t) , \tag{16}$$

where $\mathbf{u}(t)$, $\mathbf{v}(t)$ are real vectors. Combining (15) with its complex conjugate and using (16) gives first order differential equations for $\mathbf{u}(t)$ and $\mathbf{v}(t)$:

$$\dot{\mathbf{u}}(t) = A(t)\mathbf{u}(t) , \quad \mathbf{u}(-\infty) = \mathbf{d}(-\infty) , \tag{17a}$$

$$\dot{\mathbf{v}}(t) = A(t)\mathbf{v}(t) , \quad \mathbf{v}(-\infty) = \mathbf{0} , \tag{17b}$$

where $\mathbf{u}(t) = \frac{1}{2}(\mathbf{d}(t) + \mathbf{d}^*(t))$ and $\mathbf{v}(t) = \frac{1}{2}(\mathbf{d}(t) - \mathbf{d}^*(t))/i$ are real vectors for all times t and $\mathbf{d}^*$ denotes the complex conjugate of $\mathbf{d}$. If we assume reasonable restrictions on the "smoothness" of $A(t)$, unique solutions exist for (15), (17a), and (17b) [19]. The unique solution to (17b) is clearly $\mathbf{v}(t) = \mathbf{0}$, so that $\mathbf{d}(t) = \mathbf{u}(t)$ is the unique solution of (15). Thus the $d_i(t)$ are real for all times t and (13) is verified.

For an initial atomic wave function before the pulse

$$|\varphi(-\infty)\rangle = \begin{bmatrix} 1 \\ 0 \\ 0 \end{bmatrix}, \tag{18}$$

the wave function at any later time t may be written as

$$|\varphi(t)\rangle = \begin{bmatrix} d_1(t) \\ id_2(t)e^{i\phi_1} \\ d_3(t)e^{-i(\phi_2-\phi_1)} \end{bmatrix} . \tag{19}$$

$|\varphi(t)\rangle$ has a fixed phase relationship between its components that depend only on the relative phase between the two lasers, and is independent of the laser intensities or pulse shapes (the relative phase between the two lasers is fixed by either locking two independent lasers as in Refs. [3] and [4] or by generating ω_2 from ω_1 with an electro-optic modulator as in Refs. [2] and [14]). By an analogous calculation we obtain the phase behavior at any time t for an initial wave function with only population in state $|3\rangle$ before the pulse. Using the superposition principle, the result for an arbitrary superposition of initial states $|1\rangle$ and $|3\rangle$ may be deduced.

For laser pulses acting as a beam splitter with an initial wave function

$$|\varphi(-\infty)\rangle = \begin{bmatrix} a \\ 0 \\ be^{i\theta} \end{bmatrix}, \tag{20}$$

we obtain

$$|\varphi(t)\rangle = \begin{bmatrix} au_{11}(t) + bu_{31}(t)e^{i(\theta+(\phi_2-\phi_1))} \\ 0 \\ au_{13}(t)e^{-i(\phi_2-\phi_1)} + bu_{33}(t)e^{i\theta} \end{bmatrix}, \tag{21}$$

where all coefficients $u_{ij}(t)$ are real. The relative phases between u_{11}, u_{31} and u_{13}, u_{33} are independent of the laser intensity and pulse shape, even if the transfer of populations has a significant nonadiabatic component.

We have performed this calculation using the Bloch equation for the three-level density matrix and obtained similar results.

We can also understand the absence of an ac Stark shift in the dressed picture. The nonadiabatic populations of the two coupled states during the laser pulses are equal and since the dressed state eigenenergies $\pm\sqrt{(\Omega_{\text{eff}}/2)^2 - (\Gamma/4)^2}$ are symmetrically shifted about zero energy; the average shift is zero.

IV. NUMERICAL CALCULATIONS

The calculation of the transfer efficiencies and the sensitivity of result to detuning from the one-photon resonance was done by numerically integrating the interaction Schrödinger equation. For simplicity, we have restricted ourselves to the case $\Omega_{1,\max} = \Omega_{2,\max}$ and mainly considered population transfer ("atomic mirrors"). Using laser pulse shapes as indicated in Fig. 2, we obtained the coherent transfer efficiencies for the three-level system. The dashed lines in Fig. 4 show the transfer efficiencies

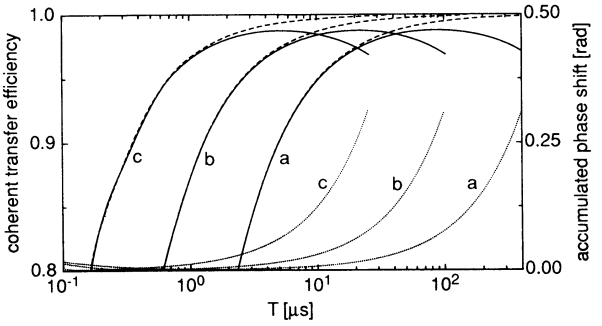

FIG. 4. Coherent transfer efficiencies (solid) and accumulated phase due to ac Stark shift (dotted) as a function of pulse length T for population transfer by adiabatic following [cesium $6S_{1/2}(F=3)$, $m_F=0 \leftrightarrow 6S_{1/2}(F=4)$, $m_F=0$ using $6P_{1/2}(F=4)$, $m_F=1$ as an intermediate state] for different laser intensities (a: 5, b: 20, c: 80 times saturation intensity, corresponding to 23 mW/cm^2, 90 mW/cm^2, and 180 mW/cm^2 for the cesium $D1$ line). The dashed lines show coherent transfer efficiencies as computed for a three-level system, neglecting off-resonant absorption. The accumulated phase due to ac Stark shift is equal to zero for this case.

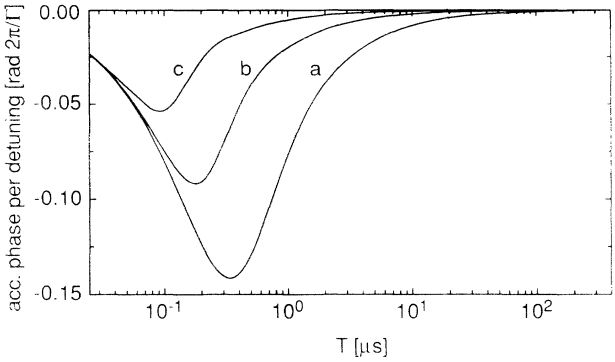

FIG. 5. Accumulated phase due to ac Stark shift during population transfer by adiabatic following in a three-level system for a detuning $\Delta=\Gamma/2\pi$ as a function of pulse length T for different laser intensities (a: 5, b: 20, c: 80 times the saturation intensity). For small detunings Δ, the accumulated phase shift is proportional to Δ.

for three different maximum laser intensities. The numerical values of the intensities are given for the magnetic field insensitive transition between the two cesium hyperfine ground states $6S_{1/2}(F=3)$, $m_F=0 \leftrightarrow 6S_{1/2}(F=4)$, $m_F=0$ using $6P_{1/2}(F=4)$, $m_F=1$ as an intermediate level ($D1$ line) and two counterpropagating lasers in a σ^+-σ^+ polarization configuration. These results have been checked by numerically integrating the Block equations.

In order to determine absolute phase shifts and the coherent part of the transfer efficiency with the Bloch equations, a fourth level not coupled to the first three levels was introduced corresponding to atoms in the other arm of the interferometer. The coherent transfer efficiency could be determined by introducing a coherence between the initial state and the fourth state before the pulse, and observing the amount of coherence that could be transferred to the final state and the fourth state. An integration of the Schrödinger equation only gives the coherent transfer efficiency.

We now discuss the effect of a small detuning Δ from the one-photon resonance. Since the dark state is still a precise eigenstate of the system with eigenvalue zero for a perfectly adiabatic process [7], there is still no accumulated phase shift. However, for a partially nonadiabatic transfer a phase shift is introduced since a detuning Δ implies that the eigenvalues of the two coupled states are no longer equally shifted from the dark state with eigenvalue zero.

The accumulated phase due to ac Stark shift is proportional to the detuning Δ for $\Delta \ll \Omega_{i,\max}$. The phase shift per detuning is shown in Fig. 5 as a function of the interaction time T for different laser intensities. The effect of detuning δ from the two-photon resonance is not expected to be a problem, since the difference can be controlled with radio frequency precision.

In real atoms, off-resonant excitation into other levels has to be taken into account. The effect of additional levels limits the transfer efficiency and introduces phase shifts due to the ac Stark effect. We have taken into account the effect of the neighboring $6P_{1/2}(F=3)$ level of the cesium $D1$ line and also the effect of the small ~ 9.2 GHz off-resonant coupling of ω_1 with the transition $|2\rangle$ to $|3\rangle$ and ω_2 with $|1\rangle$ to $|2\rangle$ by numerical integration of the Schrödinger equation of the four-level system. The expected transfer efficiencies as a function of pulse length are shown in Fig. 4 (solid lines) for different laser intensities. The maximum transfer efficiency reaches about 98.7% efficiency (independent of the laser power for $I_{i,\max} \gg I_{\text{sat}}$) for an optimum pulse length of $T \approx 500\ \mu\text{s} \times I_{\text{sat}}/I_{i,\max}$ for $I_{1,\max}=I_{2,\max}$, where $I_{i,\max}$ denotes the maximum intensity of ω_i and I_{sat} the saturation intensity [20]. The pulse length necessary to obtain a certain transfer efficiency decreases with more available laser power.

The dotted lines in Fig. 4 show the accumulated phase due to ac Stark shift during the laser pulses for the same intensities. Since the accumulated phase shift for a reasonable adiabatic process is roughly proportional to the pulse length at a given laser intensity, one might be tempted to shorten the pulse length below that required to obtain optimum transfer efficiency in order to obtain a lower phase shift. The pulse shape as plotted in Fig. 2 has been crudely optimized to match both conditions.

It should be noted that for a two-photon (e.g., Raman) pulse, where both lasers are pulsed on simultaneously and detuned far from the intermediate state to avoid spontaneous emission, the ac Stark shift of each level in general is of the order of the Rabi frequency. This shift can be canceled to first order by choosing proper detuning from the intermediate state and intensity ratio of the two beams [4].

Cancellation of the residual ac Stark shift is also possible for adiabatic population transfer is a four-level system if the lasers are detuned slightly from the one-photon resonance. The necessary detuning, which may be a few

linewidths of the excited state, can be approximately determined considering both Fig. 5 (approximately also valid for the four-level system) and Fig. 4 (dotted lines). This optimum detuning is a function of both laser pulse length and intensity. Thus compensation of the residual ac Stark shift here is more problematic and thus probably less effective than for the case of Raman π and $\pi/2$ pulses, which depends only on the ratio of the intensity of the two Raman lasers [4]. On the other hand, the phase shift accumulated during the adiabatic passage is only about 0.02 rad for 98% transfer efficiency, two orders of magnitude below the shift of a ground state level during a Raman pulse.

In a recent measurement of the photon recoil energy based on atomic interferometry using stimulated Raman transitions [3,4] ac Stark shifts differences between the two interferometers of opposite recoil shift caused by wave-front imperfections were one of the two main sources of systematic errors. This error and also phase errors directly due to wave-front imperfections were estimated to be of the same order of magnitude. These effects, although hard to quantify since the wave front errors are not accurately known, were believed to be the main cause of the eight parts in 10^{-7} (corresponding to roughly 10^{-3} rad for the final $\pi/2$ pulse) disagreement between the experimental and accepted value for the photon recoil [4]. It should be stressed that for ideal plane optical wave fronts, both for Raman transitions and adiabatic passage, errors due to ac Stark shifts cancel when the difference between both recoil components is measured. For realistic nonideal wave fronts, however, an interferometer based on adiabatic passage may yield lower systematic errors.

V. CONCLUSIONS

Coherent population transfer by delayed laser pulses seems to be an attractive method to realize high efficiency "optical elements" for atoms with small accumulated phase shifts from the ac Stark effect. We have shown that in a pure three-level system, by tuning directly on resonance with the intermediate state, two-photon resonances may be observed without any ac Stark shifts.

A possible application closely related [17] to beam splitters in atomic interferometers is an optical two-photon Ramsey experiment for high resolution spectroscopy, where the two photons are tuned on (or close to) an intermediate state. Spontaneous emission can be avoided even for a short lived intermediate state, when adiabatic following is used in each interaction region. One of the major drawbacks of two-photon spectroscopy, the ac Stark shift, can be avoided, provided no other levels are close compared to the Rabi frequency. An optical Ramsey setup based on adiabatic following thus is a promising scheme for a future extremely precise optical clock. Suitable transitions are, for example, the Tl $6P_{1/2}$-$6P_{3/2}$ and the Ba $6S^2$-$6S$ $5D$ transitions, using $7S_{1/2}$ and $6S$ $6P$, respectively as intermediate levels. For both atoms the intermediate level is more energetic than both initial and final states, so that the level scheme is similar to that in Fig. 1, except the splitting between initial and final states is in the optical regime. Both atoms have a metastable final level connected by allowed single photon transitions through the intermediate state and thus do not require high laser power.

In general, two photons of unequal frequencies are necessary to be resonant with an intermediate state. A nonzero first order Doppler shift is introduced, which requires four instead of two interaction regions, analogous to an optical one-photon Ramsey setup [21]. A pulse sequence as shown in Fig. 3 may be used for an optical two-photon Ramsey setup based on adiabatic following. Since adiabatic following is not very frequency selective a large transverse velocity spread can be used in an atomic interferometer (or optical Ramsey setup) using this method.

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[20] If we now, instead of the pulse shapes as shown in Fig. 2, assume for the pulses two displaced Gaussian shapes:

$$\Omega_1(t)=\Omega_{1,\max}\exp[-((t/T)+\tau)^2] ,$$

$$\Omega_2(t)=\Omega_{2,\max}\exp[-((t/T)-\tau)^2] ,$$

we obtain maximum transfer efficiency of 98.7% for $\tau\approx 0.5$, $T\approx 350$ $\mu s\times I_{\rm sat}/I_{i,\max}$ (cesium $D1$ line; $I_{1,\max}=I_{2,\max}$). These pulse shapes are seen by the atoms when using two spatially overlapping laser beams in an atomic beam experiment. Note that here it is less straightforward to implement pulse shapes necessary for an atomic interferometer as shown in Fig. 3.

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