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Self-induced adiabatic rapid passage

D. Grischkowsky* and M. M. T. Loy
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
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We show that, by virtue of the linewidth reduction provided by counter-propagating beams, it is possible to use the optical Stark shift to sweep a two-photon resonance through the sum frequency of the applied light while simultaneously satisfying the conditions required for optical adiabatic rapid passage. Consequently, the application of a near-resonant pulse of light can result in complete population inversion of a two-photon transition.

Two-photon resonant effects have recently attracted much attention. First, the application of the theory of single-photon coherent optics to the two-photon resonance has been of considerable interest,[1] and the observation of two-photon self-induced transparency has been reported.[2] Second, the possibility of eliminating the inhomogeneous linewidth due to Doppler broadening using two counter-propagating beams has been proposed,[3] and observed by many different laboratories.[4] Third, the recent observation of the optical Stark shift has demonstrated the importance of nonlinear effects for precision two-photon spectroscopy.[5]

In this paper we synthesize from previous work a result unique to the two-photon transition: self-induced adiabatic rapid passage. The special feature of two-photon resonance that makes this effect possible is that the optical Stark effect can shift the two-photon resonance through the sum frequency of the exciting light. By contrast, in the single-photon case the resonance is always shifted away from the frequency of the applied light. We introduce the vector model used to discuss two-photon coherent effects[1] and derive the required conditions for two-photon adiabatic rapid passage (ARP). We show that, by virtue of the linewidth reduction provided by counter-propagating beams, it is possible to use the optical Stark shift to sweep the two-photon resonance through the sum frequency of the applied light while simultaneously satisfying the two-photon ARP conditions, thereby completely inverting the two-photon transition.

The Hamiltonian describing the atom interacting with the applied light is assumed to be

$$\mathcal{H} = \mathcal{H}_0 - \vec{p}\cdot\vec{E}\,, \tag{1}$$

where $\mathcal{H}_0$ is the atomic Hamiltonian, $\vec{p}$ is the electric-dipole-moment operator, and $\vec{E}$ is the electric field of the light. The atom is irradiated by two linearly polarized light beams with frequencies ω_1 and ω_2, electric field strengths $\mathcal{E}_1$ and $\mathcal{E}_2$ and propagation vectors k_1 and k_2, respectively, i.e.,

$$\vec{E} = \hat{x}[\mathcal{E}_1\cos(k_1 z - \omega_1 t) + \mathcal{E}_2\cos(k_2 z + \omega_2 t)]\,. \tag{2}$$

We consider the two-photon transition with angular frequency Ω_{12} between the excited state $|1\rangle$ and the ground state $|2\rangle$ of $\mathcal{H}_0$. The matrix element of $\vec{p}$ between states $|1\rangle$ and $|2\rangle$ is zero.

It is possible to describe accurately the interaction of near-resonant light ($\omega_1 + \omega_2 \approx \Omega_{12}$) with the two-photon transition in the following way. The applied light induces the states $|1\rangle$ and $|2\rangle$ to evolve into the new mixed states

$$|1_m\rangle = \sum_n a_{n1}|n\rangle \tag{3a}$$

and

$$|2_m\rangle = \sum_n a_{n2}|n\rangle\,, \tag{3b}$$

where the summation index n runs over all states $|n\rangle$ of $\mathcal{H}_0$, the expansion coefficients a_{n1} and a_{n2} are obtained from time-dependent perturbation theory, and a_{11} and a_{22} are approximately equal to unity. *Now, there is a matrix element of* $\vec{p}$ *between* $|1_m\rangle$ *and* $|2_m\rangle$, *and the atom can be treated as a two-level atom in terms of these states.* Then, the vector model of Feynman, Vernon, and Hellwarth[6] can be applied to this two-level system, and the atomic response to the light is described by the precession of their unit vector $\vec{r}$ about the $\vec{\gamma}$ vector ($\vec{\gamma}=\vec{\omega}$ of Ref. 6),

$$\frac{d\vec{r}}{dt}=\vec{\gamma}\times\vec{r}\,. \tag{4}$$

The formalism therefore becomes equivalent to that of spin resonance and single-photon coherent optics. In the "rotating coordinate frame" the components of $\vec{\gamma}$ are

$$\gamma_1=-\kappa\mathcal{E}_1\mathcal{E}_2\,, \tag{5a}$$

$$\gamma_2=0\,, \tag{5b}$$

$$\gamma_3=\Omega_{12}-(\omega_1+\omega_2)+(\Delta E_1-\Delta E_2)/\hbar\,, \tag{5c}$$

where

$$\kappa=\frac{1}{2\hbar^2}\left|\sum_n p_{1n}p_{n2}\left(\frac{1}{\Omega_{n2}-\omega_1}+\frac{1}{\Omega_{n2}-\omega_2}\right)\right|\,, \tag{6}$$

and the energy shift for $|1_m\rangle$ is

$$\Delta E_1=-\sum_n\frac{p_{1n}p_{n1}\,\Omega_{n1}}{2\hbar}\left(\frac{\mathcal{E}_1^2}{\Omega_{n1}^2-\omega_1^2}+\frac{\mathcal{E}_2^2}{\Omega_{n1}^2-\omega_2^2}\right); \tag{7}$$

ΔE_2 is similarly defined. $p_{1n}=\langle 1|\,p_x\,|n\rangle$; $\Omega_{n1}=(E_n-E_1)/\hbar$, where E_n is the energy of state $|n\rangle$. The two-photon Rabi precession frequency is given by $\kappa\mathcal{E}_1\mathcal{E}_2$.[1,2] The angular frequency offset from the two-photon resonance, including the optical Stark shifts[1,5] ΔE_1 and ΔE_2 of the mixed states $|1_m\rangle$ and $|2_m\rangle$, is given by γ_3.

In terms of the vector model the requirements common to both one- and two-photon ARP are[7] (i) the angular frequency offset γ_3 must be swept through the linewidth of the transition in a time short compared to the homogeneous relaxation time, and (ii) the rate of change of γ_3 must be adiabatic, i.e.,

$$\left|\frac{d\gamma_3}{dt}\right|<\gamma_1^2\,. \tag{8}$$

In one-photon ARP experiments,[8] the frequency sweep of either the two-level system or the input laser must be obtained by some external means, such as by the dc-Stark effect or Zeeman effect of the atomic transition, or by a large frequency modulation of the laser. Here we show that in the two-photon case, the ac-Stark effect caused by the input laser fields can provide this needed frequency shift; consequently the population inversion can be achieved without any additional frequency modulation. *This self-induced ARP process is unique to the two-photon case and does not have a one-photon analog.*

For the many practical cases where resonant enhancement is used, only one intermediate state is important, and Eq. (7) for the ac Stark effect is reduced to a single term. This is the situation for the level scheme shown in Fig. 1, where $\Omega_{12}-(\omega_1+\omega_2)=\delta$ and $(\omega_2-\Omega_{n2})=\Delta$. Because the offset δ from the two-photon resonance is negligible compared to the offset Δ from the intermediate state, $(\Omega_{1n}-\omega_1)\approx\Delta$; the total frequency offset is then to a good approximation given by

$$\gamma_3=\delta+(|\,p_{1n}\mathcal{E}_1|^2-|\,p_{2n}\mathcal{E}_2|^2)/4\hbar^2\Delta\,. \tag{9}$$

For simplicity $\mathcal{E}_2$ is assumed to be the maximum field of a long pulse with rise-time τ, and $\mathcal{E}_1$ is assumed to be constant during the application of $\mathcal{E}_2$. As shown schematically in Fig. 1(b), the first ARP condition requires that the time-dependent Stark shift due to $\mathcal{E}_2$ be approximately 2δ, which is much larger than the linewidth $\delta\Omega_{12}$ of the two-

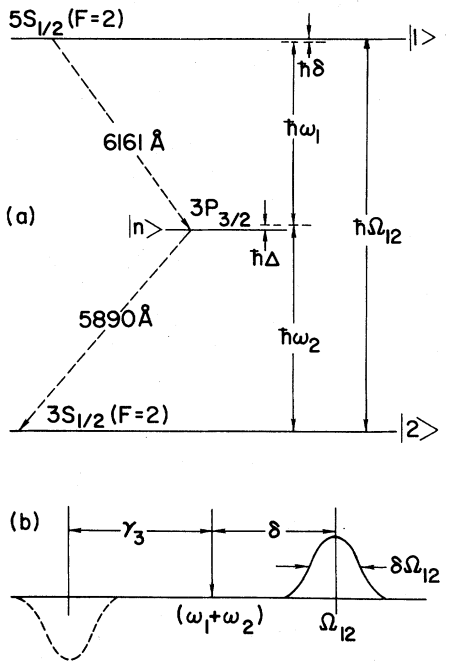

FIG. 1. (a) Energy levels of Na relevant to the two-photon transition. (b) Simplified diagram showing the initial frequency offset δ and the final offset γ_3 after application of $\mathcal{E}_2$. The two-photon resonance (solid line) is shown before the application of $\mathcal{E}_2$ and after the application of $\mathcal{E}_2$, where the resonance line (dashed line) has been completely inverted by the self-induced adiabatic rapid passage.

photon transition. It also requires that τ be less than the homogeneous relaxation time. We note that $\delta\Omega_{12}$ can be made much smaller than the Doppler width by having the two input fields propagate in opposite directions. The resulting $\delta\Omega_{12}$ will be either the residual Doppler width or the power broadened linewidth given simply by $|\gamma_1| = \kappa\mathcal{E}_1\mathcal{E}_2$. Hence, the time-dependent frequency shift required for two-photon ARP can be induced using modest field strengths. The second ARP condition can be shown to be

$$\tau > \hbar^2\Delta/|p_{1n}\mathcal{E}_1|^2 . \tag{10}$$

The application of $\mathcal{E}_2$, according to the above ARP conditions, will put all the atoms in the mixed state $|1_m\rangle$. To obtain the resulting population distribution in terms of the unperturbed states, the mixing coefficients a_{1n} must be evaluated, since the probability of occupying the intermediate state $|n\rangle$ is $|a_{1n}|^2$. For this case with only one intermediate state, $|a_{1n}|^2 = |p_{n1}|\mathcal{E}_1/2\hbar\Delta$, and the probability for occupying the unperturbed excited state $|1\rangle$ is $(1 - |a_{1n}|^2)$.

For the example of Fig. 1, the levels 1, n, and 2 are the $5S_{1/2}(F=2)$, $3P_{3/2}$, and $3S_{1/2}(F=2)$ states of Na, respectively.[9] For linearly polarized light the dipole moments are $p_{1n} = 1.1\times10^{-18}$ esu and $p_{2n} = 5.2\times10^{-18}$ esu. The transition wavelengths $\lambda_1 = 6161$ Å and $\lambda_2 = 5890$ Å are conveniently in the range of the Rhodamine 6G dye lasers. Let the input laser field strengths be $\mathcal{E}_1 = 13$ esu and $\mathcal{E}_2 = 30$ esu corresponding to the intensities of 20 kW/cm^2 and 110 kW/cm^2, respectively. To minimize single photon transitions to the intermediate state, we choose ω_1 and ω_2 such that $\Delta/2\pi c = 9$ cm^{-1} and $\delta/2\pi c = 0.01$ cm^{-1}. These values give a time-dependent frequency sweep of 500 MHz, much larger than the power broadened linewidth $\delta\Omega_{12}/2\pi = 100$ MHz. Thus, the first ARP condition will be satisfied if τ is less than the homogeneous relaxation time of the $5S_{1/2}$ state, which for low number densities is the 80 nsec radiative lifetime. The second ARP condition (Eq. 10) requires that τ be larger than 9 nsec.[10] For these experimental parameters $|a_{1n}| = 4\times10^{-3}$. Thus, to very good accuracy, the system can be considered to be completely inverted by the passage with respect to the unperturbed $5S_{1/2}(F=2)$ and $3S_{1/2}(F=2)$ states of Na; mathematically speaking, after a perfect passage the probability of finding an atom in the intermediate state is only 1.6×10^{-5}. The mixing coefficients also allow us to estimate the probability of excitation to the 5S state by the two-step process where the atom is first excited to the 3P state and from there to the 5S state. For this example, the probability for this process is of the order of $|a_{1n}|^2|a_{2n}|^2$ and is completely negligible.

In order to demonstrate experimentally that self-induced adiabatic rapid passage under the above conditions does indeed transfer all the sodium atoms from the $3S_{1/2}(F=2)$ ground state to the $5S_{1/2}(F=2)$ excited state, the number of atoms in the $F=2$ ground state can be continuously monitored during the passage. This can be conveniently done by tuning a very low power (to avoid any saturation effects) cw laser $\mathcal{E}_3$ to the transition from the $F=2$ ground state to the $3P_{1/2}$ state; the absorption from the $F=1$ ground state to the $3P_{1/2}$ state will be negligible. Here the Stark shift of the ground state caused by the application of $\mathcal{E}_2$ is relatively small compared to the Doppler width. This technique has the advantage of requiring very low atomic-number densities, and thereby eliminates any propagation effects associated with beams $\mathcal{E}_1$ and $\mathcal{E}_2$. The number density N need only be sufficient to attenuate beam $\mathcal{E}_3$ by a reasonable amount such as e^{-1}, which for a 1-cm cell requires $N = 3.2\times10^{11}$ atoms/cm^3. After the two-photon ARP the absorption of beam $\mathcal{E}_3$ will be negligible.

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[9]Because of the two-photon selection rule of $\Delta F = 0$, the nearest line is 1600 MHz away and can be neglected.

[10]The $3P_{1/2}$ state is 17 cm^{-1} below the $3P_{3/2}$ state. In this example, inclusion of the $3P_{1/2}$ state in the calculation changes the numbers slightly, but the ARP conditions remain satisfied.