Efficient Population Transfer in a Three-Level Ladder System by Frequency-Swept Ultrashort Laser Pulses

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Enhanced population transfer from the lowest to the highest level in the three-state ladder $5s \rightarrow 5p \rightarrow 5d$ of the rubidium atom by frequency-swept ultrashort laser pulses is observed if the frequency sweep first makes the pulse resonant with the $5s \rightarrow 5p$ transition, and later with the $5p \rightarrow 5d$ transition. With this "intuitive" scheme it is possible to achieve complete population inversion via the intermediate level by two successive adiabatic passages. Complete inversion is also obtained by a "counterintuitive" direction of the frequency sweep, in which case population of the intermediate level during the pulse is strongly reduced.

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Efficient preparation of an ensemble of atoms or molecules in one specific state is of considerable interest not only in spectroscopy, but also in studies of chemical dynamics. Recently, Chelkowski, Bandrauk, and Corkum [1] discussed a novel method for efficient, controlled molecular dissociation using ultrashort (10^{-14}–10^{-11} s) frequency-swept ("chirped") laser pulses. For the case of a diatomic molecule they suggested constructing a laser pulse with a frequency sweep such that the frequency follows the vibrational level spacing. Such a pulse makes the wave function of the nuclei of the molecule "climb up" the vibrational ladder, finally leading to greatly enhanced dissociation as compared to a chirp-free pulse of the same intensity. In this Letter we report on the role of a frequency sweep on such an ultrashort pulse in an atomic rather than a molecular ladder. In the $5s \rightarrow 5p \rightarrow 5d$ sequence (hereafter also denoted by 1→2→3) in the rubidium atom [Fig. 1(a)], we measured the population of the upper ($5d$) level after excitation with a frequency-swept laser pulse. It is observed that at relatively low intensities this population is greatly enhanced if the laser frequency is resonant with the first step of the ladder (1→2) in the beginning of the pulse, and with the second step (2→3) at the end. With such an intuitively correct frequency sweep, 100% of the population can be transferred to the upper state, while halfway during the pulse all population resides in the intermediate 5p state. Although this process is coherent, a necessary condition for full population transfer, it is not sensitive to the precise pulse characteristics. If, in general, an intermediate state experiences severe decay, it is favorable to keep the (transient) population of this state low. We found that this can be achieved by applying a *reversed*, "counterintuitive" frequency sweep, such that the laser frequency is first resonant with the second step of the ladder, and later with the first step. In this case the maximum population in the intermediate state is considerably reduced, while still 100% is transferred to the upper state. The use of an ultrashort pulse in itself already reduces relaxation processes during excitation. This is necessary if bond-selective dissociation of polyatomic molecules is desired, as mentioned in Ref. [1], since intramolecular relaxation takes place on a picosecond time scale. In our atomic case, the duration of the pulse (10^{-13}–10^{-11} s) ensures that spontaneous decay can be neglected (lifetimes of the states are $> 10^{-8}$ s).

In the experiment, rubidium atoms in a vacuum system were exposed to an ultrashort laser pulse. The population of the upper level of the ladder ($5d$) was then probed by photoionization with a long pulse. Tunable excitation pulses with a wavelength around 778 nm and a bandwidth large enough to encompass both steps of the ladder were obtained as follows (details can be found in

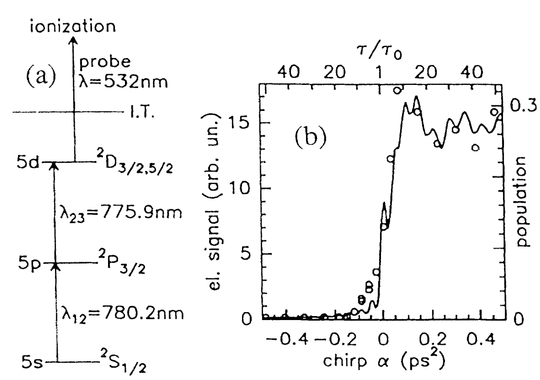

FIG. 1. (a) Energy levels of the three-state ladder in rubidium. I.T. denotes the ionization threshold. (b) Measured photoelectron signal (points) and calculated populations of the upper level (full curve) as a function of frequency sweep α (λ_c = 778.5 nm, $\Delta\lambda$ = 5.8 nm, F = 25 μJ/cm^2). Upper x axis indicates pulse duration relative to a chirp-free pulse. A red-to-blue sweep ($\alpha > 0$) transfers much more population than the blue-to-red sweep ($\alpha < 0$).

Ref. [2]): Pulses from a colliding-pulse mode-locked dye laser were amplified at 10 Hz in four dye cells pumped by the second harmonic of a Nd:YAG (yttrium aluminum garnet) laser. These pulses were focused in water to generate a wavelength continuum, which was led through a pulse shaper [3]. This powerful device selects both central wavelength and bandwidth of the square-shaped power spectrum. In addition, one can make the path length through the shaper wavelength dependent, thereby controlling the chirp continuously. The pulses were then amplified again to an energy of $\sim 10\ \mu$J. To avoid spatial intensity inhomogeneities, the beam (diameter 2.5 mm) remained unfocused. It was crossed with an effusive beam of rubidium atoms coming out of an oven.

After excitation, the population of the upper level was probed by one-photon ionization [Fig. 1(a)] with a pulse from the frequency-doubled (injection-seeded) Nd:YAG laser (wavelength 532 nm, duration 5 ns). The pulse energy was $\sim 800\ \mu$J and the beam diameter was 1.5 mm, somewhat smaller than that of the excitation beam in order to probe only the atoms excited by the central (constant-intensity) part of the first beam. The photoelectrons were detected with a multichannel plate detector. The wavelength of the probe pulse allowed one-photon ionization only from the upper level, and the intensity ($I = 8\times 10^6$ W/cm^2) was sufficiently low to avoid two-photon ionization from the other levels. Ionization by the excitation pulse was not observed (mainly) because of its low fluence ($\leq 500\ \mu$J/cm^2; compare to 45 mJ/cm^2 for the probe).

Before presenting the experimental results, we will first explain our measure for the amount of chirp. After the pulse shaper, the field of the pulse in the frequency domain is $E(\omega) = |E(\omega)|e^{i\alpha(\omega-\omega_0)^2}$. Here $|E(\omega)|$ denotes the square-shaped frequency content of the pulse, ω_0 the central (angular) frequency, and α the chirp. The value of α can be obtained directly from distances and angles in the pulse shaper [4]. If $\alpha = 0$ the pulse is transform limited, implying the minimal pulse duration τ_0. A red-to-blue frequency sweep corresponds to $\alpha > 0$, and a blue-to-red sweep to $\alpha < 0$. The larger the $|\alpha|$, the longer the pulse duration τ. A good approximation for the relative lengthening is [4] $\tau/\tau_0 \approx \sqrt{1+(\frac{|\alpha|(\Delta\omega)^2}{\pi})^2}$, where $\Delta\omega$ denotes the bandwidth of the pulse.

Figure 1(b) shows a typical result of the measured photoelectron signal from the upper state at relatively low (constant) energy per pulse (fluence $F = 25\ \mu$J/cm^2). Central wavelength ($\lambda_c = 778.5$ nm) and bandwidth ($\Delta\lambda = 5.8$ nm) were chosen such that both λ_{12} (780.2 nm) and λ_{23} (775.9 nm) were within the spectrum of the pulse. The corresponding bandwidth-limited pulse duration was $\tau_0 = 310$ fs. It is clear from Fig. 1(b) that the intuitively correct red-to-blue sweep ($\alpha > 0$) transfers population much more efficiently than the blue-to-red sweep. It is also more effective than a chirp-free pulse, even though the latter has a higher intensity, since chirp lengthens the pulse and the pulse energy was kept constant [the relative pulse lengthening τ/τ_0 is also given in Fig. 1(b)]. If the (peak) *intensity*, rather than the pulse energy, is kept constant, the difference between the frequency-swept and the bandwidth-limited pulse is much more pronounced. Such a comparison may be relevant in situations where the maximum useful intensity is limited by other competing processes, as in the case of photodissociation of molecules, where above $\sim 10^{13}$ W/cm^2 significant photoionization sets in [1].

When the pulse energy was increased by a factor of 20 as compared to Fig. 1(b), the results in Fig. 2(a) were obtained. Although these are qualitatively similar to Fig. 1(b), the most important observation here is the constant value of the measured electron signal for large enough red-to-blue sweep.

Surprisingly different behavior is found for $\lambda_c \approx 780.8$ nm [see Fig. 2(b)], and otherwise similar pulse parameters as in Fig. 2(a). In this case a counterintuitive blue-to-red frequency sweep ($\alpha < 0$) transfers population much more effectively than the red-to-blue sweep.

As a first step to explain the results we performed a numerical calculation of the population in the upper level after excitation, based on the Schrödinger equation for a three-level system without loss [5]. All relevant parameters in this calculation were obtained directly from the experiment. The results are also shown in Figs. 1 and

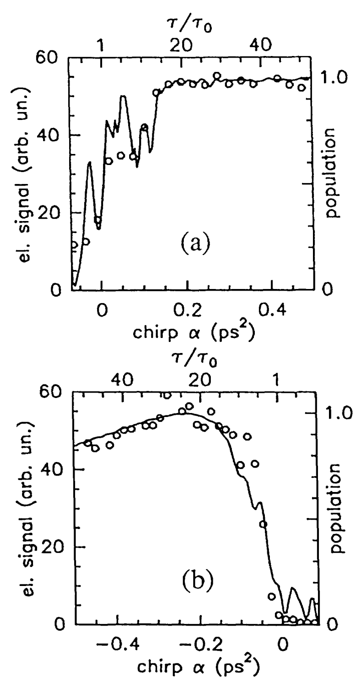

FIG. 2. (a) As in Fig. 1(b), for the following parameters: $\lambda_c = 777.5$ nm, $\Delta\lambda = 5.8$ nm, $F = 500\ \mu$J/cm^2. Full population transfer is obtained for $\alpha > 0.2$ ps^2 (red-to-blue sweep). (b) As in Fig. 1(b), for the following parameters: $\lambda_c = 780.8$ nm, $\Delta\lambda = 5.8$ nm, $F = 500\ \mu$J/cm^2. Full population transfer is obtained around $\alpha = -0.2$ ps^2 (blue-to-red sweep).

2. Once the vertical scale of one measurement was fixed to the calculated scale, every calculation fitted the corresponding measurement as well. Therefore we assume that the calculated populations have indeed been achieved in the measurements.

The right y axes in Figs. 1(b) and 2 then show that 100% population transfer can be obtained both with a red-to-blue and blue-to-red sweep. For large enough sweep this result is insensitive to the exact value of the sweep (and of other pulse parameters, as will be discussed later). This is one of the general, attractive, properties of the mechanism underlying this population transfer, which is adiabatic passage (AP) [6]. Population inversion by AP between two levels is achieved when the detuning between the frequency of the radiation field and the resonance frequency of the system is made to change sign slowly enough to pass the system adiabatically through the region of avoided crossing, leaving it in the same dressed state at all times. This can be done either by changing the resonance frequency (as in the first observation of AP [7]) or by modulating the laser frequency, as was done for nanosecond laser pulses [8]. Very recently, AP by chirping of picosecond pulses has been used to create inversion in an ensemble of molecules [9]. It has also been shown that the large bandwidth of such ultrashort pulses does not necessarily imply loss of selectivity [10]. The advantages of AP for obtaining population inversion, as compared to π pulses, are well known (see, e.g., [11]). Here we only mention that AP is insensitive to the exact pulse parameters and the precise frequency of the resonance, and therefore can be used to invert an inhomogeneously broadened ensemble of particles.

In the case of the intuitive red-to-blue chirp, *two* sequential adiabatic passages driven by *one* frequency-swept laser pulse first transfer the population from the ground state to the intermediate level, and subsequently from the intermediate level to the upper level. This mechanism, which works as long as the adiabatic condition is satisfied for both steps, can be seen in Fig. 3(a), where the calculated dressed-atom eigenvalues of the three-step ladder are plotted as a function of frequency (at a fixed laser intensity). A sufficiently slow red-to-blue chirp adiabatically transfers population via path A. The calculated populations of the levels as a function of time [Fig. 3(b)] clearly illustrate that halfway during the pulse almost all population resides in the intermediate level. An earlier observation of a series of adiabatic passages was made with a constant driving (microwave) field and an additional swept electric field to pass successive atomic transitions through resonance with the driving field [12].

Figure 3(a) also helps to explain complete population inversion by a pulse with a counterintuitive blue-to-red frequency sweep. In this case the system adiabatically passes through the level crossing between the ground state and the upper level directly (path B). The central wavelength of the required pulse is shifted to the red due to the difference in transition dipole moment between the

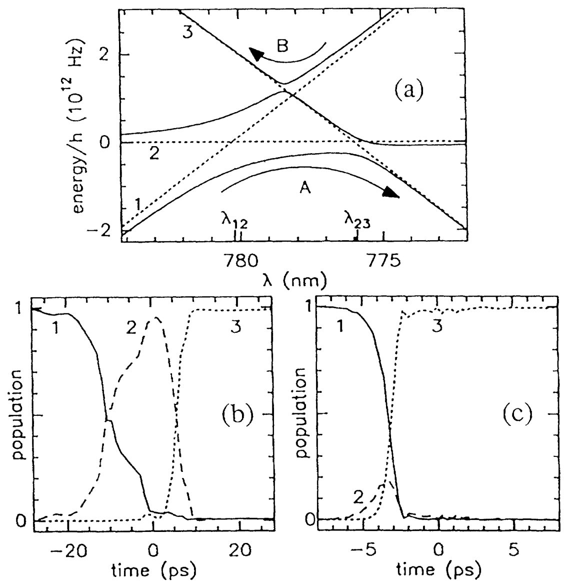

FIG. 3. (a) Calculated dressed-level scheme of the three-state ladder at an intensity $I = 1.1 \times 10^7$ W/cm^2. Levels without coupling are also shown (dotted curves): lines 1, 2, and 3 correspond to dressed states $|5s+\omega\rangle$, $|5p\rangle$, and $|5d-\omega\rangle$, respectively. Path A results from the "intuitive" red-to-blue frequency sweep; path B from the "counterintuitive" sweep. (b) Calculated time evolution of the populations of the levels resulting from the intuitive red-to-blue chirp. Halfway during the pulse almost all population resides in the intermediate level. Pulse parameters are $\lambda_c = 777.5$ nm, $\Delta\lambda = 5.8$ nm, $\alpha = 0.5$ ps^2 ($\tau/\tau_0 = 52$), $F = 500$ μJ/cm^2. (c) As in (b) for the pulse with the counterintuitive blue-to-red chirp. Population of the intermediate state is strongly reduced. Pulse parameters are $\lambda_c = 780.8$ nm, $\Delta\lambda = 5.8$ nm, $\alpha = -0.24$ ps^2 ($\tau/\tau_0 = 26$), $F = 500$ μJ/cm^2.

two steps [$\mu_{12} = 5.87$ atomic units (a.u.), $\mu_{23} = 1.77$ a.u. [13]], as was indeed observed. In the model there is no direct transition dipole moment between the ground state and the upper level. The avoided crossing between these states shows up as a consequence of the *combined* coupling between the lower and middle state, and between the middle and upper state. Therefore the counterintuitive mechanism only works well when the $5s \to 5p$ and $5p \to 5d$ transitions are sufficiently mixed, which is the case when at least one of the (peak) Rabi frequencies is comparable to (or larger than) the difference between the transition frequencies of the two steps, $\Delta\nu_{\text{trans}}$ ($= 2.1$ THz). The peak Rabi frequency for the strongest transition ($5s \to 5p$) is given by $2\pi\Omega_{R12} = \mu_{12}E/h$, where E denotes the electric field strength of the light. For the pulse parameters of Fig. 2 and $|\alpha| = 0.2$ ps^2, it is found to be $2\pi\Omega_{R12} = 2.1$ THz $= 1.0\Delta\nu_{\text{trans}}$. So the transitions are indeed sufficiently mixed. The slow leveling off of the final population in Fig. 2(b) as α drops below -0.3 ps^2 is caused by the decrease of the peak Rabi frequency as $|\alpha|$ increases ($2\pi\Omega_{R12} = 1.4$ THz

$= 0.7\Delta\nu_{\rm trans}$ at $\alpha = -0.5$ ps^2). The peak Rabi frequencies in Fig. 1(b) are a factor of 4.5 smaller than those in Fig. 2, so $2\pi\Omega_{R12} < 0.2\Delta\nu_{\rm trans}$, for $|\alpha| > 0.2$ ps^2. This shows that Fig. 1(b) corresponds to the low-intensity limit, where the $5s \rightarrow 5p$ and $5p \rightarrow 5d$ transitions are not mixed.

It is worth pointing out a relation between the counterintuitive scenario and stimulated Raman adiabatic passage (STIRAP) [14], in which population is counterintuitively inverted in a Λ system by applying two separate laser pulses: *first* the Stokes pulse and *then* (partially overlapping in time) the pump pulse. This pulse sequence prevents significant population of the intermediate state during the pulses. Recent calculations show that this idea remains valid even if the "intermediate state" is in the continuum [15]. Also in our case the counterintuitive frequency sweep reduces the population of the intermediate state during the pulse [see Fig. 3(c)], because the pulse first mixes the intermediate and upper level, thereby facilitating "direct" population of the upper state later. However, our approach to use a single, counterintuitively chirped pulse for creating population inversion by AP is not applicable to a Λ system because of the absence of a direct crossing between the dressed initial and final level.

In conclusion, we have shown experimentally that frequency-swept ultrashort laser pulses can provide efficient population transfer in ladder systems. The ideal case of 100% population transfer can be obtained by successive adiabatic passages when the frequency sweep follows the ladder spacing. This process is not sensitive to the precise pulse characteristics. Furthermore, population of intermediate states can be reduced, while keeping 100% transfer, by applying a reversed, counterintuitive frequency sweep.

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