Robust Population Transfer by Stimulated Raman Adiabatic Passage in a Pr^{3+}:Y_2SiO_5 Crystal

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We report on the experimental implementation of stimulated Raman adiabatic passage (STIRAP) in a Pr^{3+}:Y_2SiO_5 crystal. Our data provide clear and striking proof for nearly complete population inversion between hyperfine levels in the Pr^{3+} ions. The transfer efficiency was monitored by absorption spectroscopy. Time-resolved absorption measurements serve to monitor the adiabatic population dynamics during the STIRAP process. Efficient transfer is observed for negative pulse delays (STIRAP), as well as for positive delays. We identify the latter by an alternative adiabatic passage process.

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In recent decades, a large number of experimental studies in quantum information science has been conducted in atomic media in the gas phase [1]. However, also some particular solid state systems were considered as promising candidates for a successful implementation of coherent interactions between light and matter. Appropriate solid media are quantum dots [2,3], color centers in diamond [4], or dielectric crystals doped with rare-earth-metal ions (rare-earth-metal materials) [5,6].

First implementations of electromagnetically induced transparency, slow light, and light storage [7] in solids were carried out in Pr^{3+}:Y_2SiO_5 crystals (hereafter Pr:YSO) [8–10]. Pr:YSO and other rare-earth-metal materials provide excellent properties for quantum information processing. These media exhibit narrow optical linewidths and long decoherence times. The hyperfine levels of the ground state are well suited for the implementation of qubits. The permanent electric dipole moment facilitates control of the mutual interaction of qubits by means of optical excitation.

Robust quantum computing requires transfer processes insensitive to fluctuations of the experimental parameters. Rapid adiabatic passage is a robust technique for complete population transfer to an excited state [11]. Therefore, it permits efficient control of qubit-qubit interactions in rare-earth-metal-ion-doped crystals. The basic implementation of rapid adiabatic passage was recently demonstrated in these rare-earth-metal materials [12,13]. However, Raman-type transfer processes are required to directly prepare and manipulate qubits encoded in hyperfine levels of the ground state.

Stimulated Raman adiabatic passage (STIRAP) [11] provides complete coherent population transfer in a lambda-type quantum system independent of radiative losses. STIRAP is robust against correlated phase fluctuations of the driving laser fields [14]. STIRAP and its extensions [15,16] are basic tools for coherent state manipulation and preparation. Therefore, it is of great importance for applications in quantum information processing. STIRAP is the subject of a large number of investigations in the gas phase, though there is no convincing demonstration and systematic investigation of STIRAP in solids so far. Goto and Ichimura [17] reported on first evidences for STIRAP in Pr:YSO. The authors of Ref. [17] combined few data points from time-domain absorption measurements at fixed frequency, complex pulse propagation calculations, and fit procedures to deduce variations of transfer efficiencies. However, this approach did not provide a direct, convincing, and systematic measurement of STIRAP.

In the following, we report on the demonstration of STIRAP in a Pr:YSO crystal, monitored by straightforward spectroscopy. The data provide a *direct*, very clear, and systematic measure for adiabatic population transfer by STIRAP. The data indicate a transfer efficiency approaching 100%, although the laser pulses suffer from frequency jitter. This confirms the robustness of STIRAP. In addition, time-resolved absorption measurements reveal the population dynamics characteristic for STIRAP. Moreover, we also observe efficient population transfer if the pulse sequence is reversed with respect to the case of STIRAP.

For a basic theoretical description of STIRAP we consider a three-level lambda-type quantum system of bare states $|1\rangle$, $|2\rangle$, and $|3\rangle$ (see Fig. 1). Initially, all population is in state $|1\rangle$. The pump pulse excites the transition between states $|1\rangle$ and $|2\rangle$. The Stokes pulse excites the transition between states $|2\rangle$ and $|3\rangle$. The coupling strengths are defined by the Rabi frequencies $\Omega_P(t) = \mu_{12}\mathcal{E}_P(t)/\hbar$ and $\Omega_S(t) = \mu_{23}\mathcal{E}_S(t)/\hbar$, with the dipole transition moments μ_{ij} and the electric fields of the lasers $\mathcal{E}_{P,S}(t)$. We consider now the adiabatic states, i.e., the instantaneous eigenstates of the system, including the interaction with the radiation fields. If the lasers are tuned to two-photon resonance between states $|1\rangle$ and $|3\rangle$, the *dark* eigenstate of the system reads:

$$|d\rangle = \cos\theta \cdot |1\rangle - \sin\theta \cdot |3\rangle \quad (1)$$

with the time-dependent mixing angle θ given by

$$\theta(t) = \arctan[\Omega_P(t)/\Omega_S(t)]. \quad (2)$$

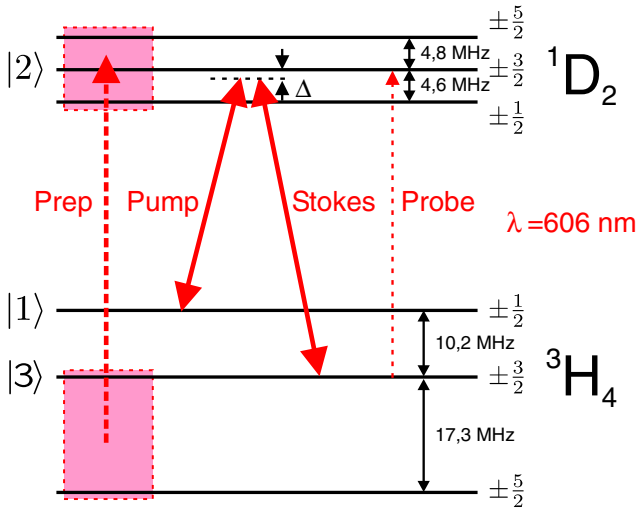

FIG. 1 (color online). Coupling scheme in Pr:YSO. Δ is the detuning of pump and Stokes laser from the single-photon resonances.

First, we consider a counterintuitive pulse sequence, i.e., the Stokes pulse precedes the pump pulse. Initially, i.e., at $t \to -\infty$, the mixing angle is $\theta = 0$. Thus $|d\rangle \equiv |1\rangle$. At the end of the interaction, i.e., at $t \to +\infty$, the angle becomes $\theta = \pi/2$. Thus $|d\rangle \equiv -|3\rangle$. Provided the evolution is adiabatic, the system remains in the dark state during the interaction. Thus, in the counterintuitive pulse sequence, all population is transferred to the target state by STIRAP. As the intermediate state $|2\rangle$ is never populated throughout the interaction, the transfer efficiency is independent of losses from this state. Moreover, phase fluctuations of the driving laser fields will only moderately affect the transfer efficiency, provided the phase fluctuations of both fields are correlated. Then the phase factors cancel out according to Eq. (2).

If the lifetime of the intermediate state $|2\rangle$ is long compared to the radiation pulses, efficient adiabatic transfer is also possible for an intuitive pulse sequence, i.e., pump pulse preceding Stokes pulse [18]. In this case, the system remains in the *bright* state

$$|b\rangle = \cos\varphi \cdot (\sin\theta \cdot |1\rangle + \cos\theta \cdot |3\rangle) - \sin\varphi \cdot |2\rangle, \quad (3)$$

with the additional time-dependent mixing angle

$$\varphi(t) = \tfrac{1}{2}\arctan[\sqrt{\Omega_P^2(t) + \Omega_S^2(t)}/\Delta]. \quad (4)$$

For $\Delta \neq 0$, i.e., if the lasers are detuned from the single-photon resonance, the angle φ is zero before and after the interaction. For the intuitive pulse sequence, the bright state evolves from state $|b\rangle \equiv |1\rangle$ towards $|b\rangle \equiv |3\rangle$. In contrast to STIRAP, the intermediate state $|2\rangle$ is populated during the transfer process. Thus, radiative losses from state $|2\rangle$ are possible and the adiabaticity of the transfer process is not maintained. This leads to a reduction of the transfer efficiency. Only if the radiative lifetime of the intermediate state $|2\rangle$ is far longer than the interaction time, efficient population transfer is possible via the bright state $|b\rangle$. We will refer to the transfer process via the bright state $|b\rangle$ as b-STIRAP.

For $\Delta = 0$, b-STIRAP breaks down and the target state population becomes sensitive to the Rabi frequencies. This leads to a reduction of the average transfer efficiency. Therefore, the population distributions presented in Ref. [17] for intuitive pulse order can be understood in the framework of b-STIRAP.

Figure 1 shows the coupling scheme for STIRAP in Pr:YSO. The pump and the Stokes laser drive transitions involving hyperfine components of states $^3H_4$ and $^1D_2$. The lifetime of the excited state is $\tau_2 = 164$ μs [19]. As the inhomogeneous broadening of the optical transitions exceeds the energy splitting of the hyperfine sublevels by 2 orders of magnitude, a single laser field can drive transitions in different ensembles of ions within the inhomogeneous bandwidth at the same time.

To selectively drive and probe an adiabatic process in a single ensemble of praseodymium ions with well-defined transition frequency, we use a preparation process based on spectral hole burning [20]. We apply a preparation laser pulse (rectangular temporal shape, pulse duration 84 ms) with frequency ν_0 initially centered in the inhomogeneous bandwidth. The frequency of the preparation pulse is repeatedly swept over a range of 0 MHz $< \Delta\nu <$ 18 MHz. Because of optical pumping, the preparation process creates a transmission window in the inhomogeneous bandwidth, i.e., a spectral range, which shows no absorption. After the preparation pulse, the pump pulse and the Stokes pulse are applied. We choose the frequencies of the pump and Stokes pulse as $\nu_P - \nu_0 = -5$ MHz and $\nu_S - \nu_0 = +5.2$ MHz. The pump frequency is outside the transmission window, i.e., the pump laser experiences absorption. The Stokes frequency is inside the transmission window, i.e., initially the Stokes laser experiences no absorption. Thus, the preparation process prepares the system in a lambda-type coupling scheme with all population initially in state $|1\rangle$, which is a necessary requirement for STIRAP. The frequency difference $\nu_S - \nu_P$ matches the difference between states $|1\rangle$ and $|3\rangle$. The population distribution in the medium is monitored via the absorption of a probe laser at frequency $\nu_{\text{Probe}} - \nu_0 = +5.2$ MHz. Initially, if all population is in state $|1\rangle$, the probe laser is transmitted without losses. When STIRAP modifies the population distribution, the absorption of the probe laser is a direct and absolute measure for the population in state $|3\rangle$.

In the experiment, a Pr:YSO crystal (length 3 mm) with a dopant concentration of 0.02% is cooled to cryogenic temperatures. A single-longitudinal mode dye laser (Coherent 699) provides radiation at 605.98 nm. The linewidth of 1 MHz results from frequency jitter. The laser radiation is split into different beam lines. Each beam is intensity modulated and frequency shifted by acousto-

optical modulators in double-pass configuration. The beam lines provide the preparation, pump, Stokes, and probe laser pulses. Therefore, phase fluctuations due to frequency jitter are identical for all laser pulses, as all beam lines are derived from a single laser source. The weak probe beam is counterpropagating with respect to the other beams. After passing the sample, a small fraction of the probe laser intensity is directed onto a silicon photodiode to monitor absorption. All beams exhibit parallel and linear polarization, chosen to yield maximum absorption in the Pr:YSO crystal. In the interaction region, the diameters (FWHM of intensity) of the preparation, pump, and Stokes beam are approximately 295 μm, while the diameter of the probe beam is 175 μm. The laser powers are approximately 100 mW for the preparation and pump beams and 65 mW for the Stokes beam. The probe beam is attenuated to 12 μW. These intensities correspond to peak Rabi frequencies $\Omega_P = 2\pi \times 703$ kHz, $\Omega_S = 2\pi \times 715$ kHz, and $\Omega_{\text{Probe}} = 2\pi \times 14$ kHz. The temporal shapes of the pump and the Stokes pulse exhibit an envelope close to a Gaussian with a duration (FWHM of intensity) of $\tau_P = 19.8$ μs and $\tau_S = 24.6$ μs. The delay of the pump and Stokes pulse is variable. The probe pulse exhibits a rectangular shape with a pulse duration of $\tau_{\text{Probe}} = 20$ μs. The probe pulse is delayed by 600 μs with respect to the pump pulse. The preparation pulse sequence is well delayed prior to the other laser pulses.

In the experiment, the absolute transmission of the probe laser pulse is measured. Application of Beer's law permits calculation of the absorption coefficient. The change of this absorption coefficient induced by the two driving laser fields is proportional to the net population transfer to the target state $|3\rangle$. As this state is initially not populated due to the repetitive preparation process, only well-known spectroscopic constants of Pr:YSO [20] are required for the calibration of population transfer efficiency. Thus, this simple and straightforward method permits direct observation of population redistributions, e.g., driven by STIRAP.

In a first experiment, we observed the variation of the transfer efficiency versus the detuning from the two-photon resonance between states $|1\rangle$ and $|3\rangle$ (see Fig. 2). In the experiment, the pump laser frequency was kept fixed, while the Stokes laser frequency was varied. When the Stokes laser frequency is detuned from the resonance, population transfer to the target state is due to optical pumping by the pump pulse alone. A low transfer efficiency of $(15 \pm 5)\%$ is observed. However, if the Stokes laser pulse is tuned to match the resonance, the transfer efficiency increases strongly as the system is driven in a Raman process. For coincident laser pulses the transfer reaches a maximum of 50%. The data (solid triangles) show an asymmetry with respect to the two-photon resonance and are well described by a Fano profile (dashed line). This can be understood in terms of coherent population trapping detuned from single-photon resonance [21].

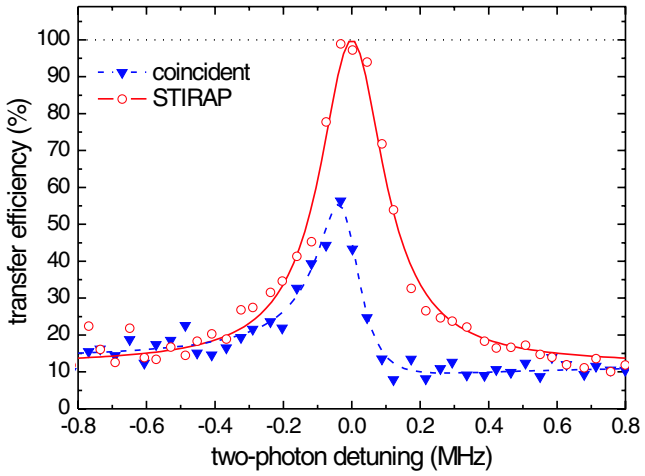

FIG. 2 (color online). Transfer efficiency versus two-photon detuning. The pulse delay in the case of STIRAP was -15 μs (Stokes preceding pump). Lines are fit functions to improve visibility.

For delayed laser pulses with counterintuitive pulse sequence, i.e., STIRAP, the experimentally determined transfer efficiency approaches 100%. Thus, we observe nearly complete population transfer.

In a second experiment, time-resolved absorption measurements served to monitor the adiabatic population dynamics of STIRAP (see Fig. 3). For this measurement, we reduced the duration of the probe pulse to 5 μs to increase temporal resolution. The two-photon resonance was maintained throughout the measurement. We varied the delay of the probe laser pulse. Negative delay corresponds to the probe laser pulse preceding pump and Stokes pulse. For negative delay the probe pulse experiences, as expected, no absorption. For delays towards positive values, the probe pulse follows the pump and Stokes pulse. Now, the attenu-

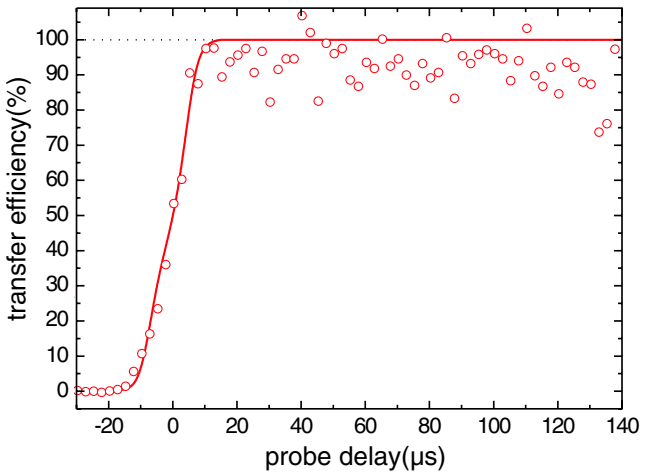

FIG. 3 (color online). Adiabatic population dynamics of STIRAP observed by time-resolved absorption measurements. The solid line is a theoretical *ab initio* prediction.

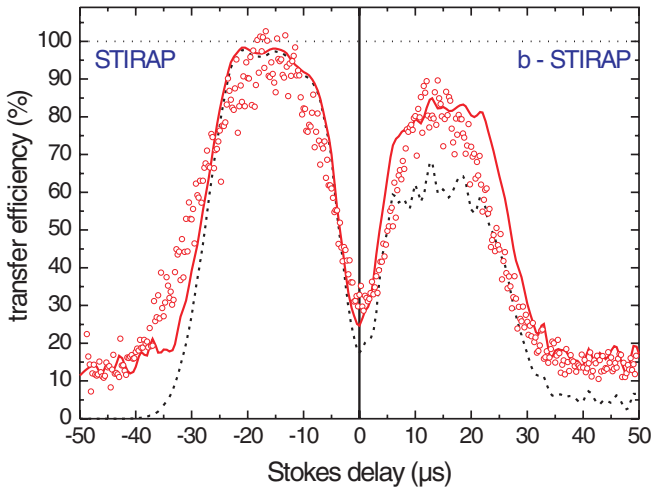

FIG. 4 (color online). Transfer efficiency vs delay of the Stokes pulse with respect to the pump pulse. Data agree well with numerical simulation (solid line) including statistical frequency jitter [(Gaussian distribution with bandwidth 1 MHz (FWHM)] assuming an average detuning $\Delta = 2\pi \times 320$ kHz. Results excluding the contribution of incoherent population transfer are shown for comparison (broken line).

ation of the probe laser pulse increases rapidly as STIRAP populates the target state. The transfer efficiency approaches 100%. The data agree well with an *ab initio* calculation (solid line).

In a third experiment, we monitored the transfer efficiency versus the time delay between the Stokes and the pump pulse (see Fig. 4). The probe pulse is again delayed by 600 μs with respect to the pump pulse. We observe transfer efficiencies approaching unity for negative pulse delays (STIRAP) and positive pulse delays (*b*-STIRAP). In both cases, the transfer efficiency is far larger than in the case of coincident laser pulses. For delays larger than the pulse durations, i.e., no considerable temporal overlap between the laser pulses, the transfer efficiency approaches the limit of optical pumping by the pump pulse. The maxima in the transfer efficiency show prominent plateaus, in particular, for the case of negative pulse delays (STIRAP). Thus, the transfer efficiency does not vary over an extended range of delays. Already this feature is a clear evidence for robust, adiabatic population transfer. The population transfer is nearly complete, i.e., approaches unity, for negative delays (STIRAP). As expected, in the case of an intuitive pulse sequence (*b*-STIRAP) the maximum transfer efficiency is reduced. Experimental data are well reproduced by numerical simulations including frequency jitter. Comparison of the results including and excluding incoherent population transfer (see Fig. 4) shows that for STIRAP the population transfer is fully coherent.

However, for *b*-STIRAP an incoherent contribution to the transfer efficiency exists. Because of frequency jitter the *b*-STIRAP process is imperfect and leaves the intermediate state partially populated after the interaction.

In conclusion, we have demonstrated robust and efficient population transfer by STIRAP in a Pr:YSO crystal. We investigated (i) the variation of the transfer efficiency versus the laser frequencies, (ii) the temporal evolution of the population of the target state, and (iii) the variation of the transfer efficiency versus the time delay between the driving laser pulses. We observed efficient, adiabatic population transfer both for negative (STIRAP) as well as for positive delay (*b*-STIRAP). All our data and the systematic measurements are striking proofs for population inversion driven by STIRAP in a solid.

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