Confining atomic populations in space via stimulated Raman adiabatic passage in a doped solid

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Abstract
We experimentally demonstrate spatial confinement of atomic excitation by adiabatic passage processes in a rare-earth ion-doped \textit{Pr}^{3+}:Y_2SiO_5 crystal. In particular, we apply stimulated Raman adiabatic passage (STIRAP) and compare its performance with electromagnetically induced transparency (EIT). Using a Stokes beam with Gaussian and a pump beam with donut shape we localize the atomic population in the zero-intensity center of the latter. Our data confirm that adiabatic passage confines excitation far below the diameter of the driving laser beams, and that this localization rapidly increases with laser intensity. We find, that STIRAP significantly outperforms EIT, as it was predicted by previous theory proposals, i.e., STIRAP reaches small excitation volumes with much lower laser intensity. The experimental data agree very well with numerical simulations. The findings serve as a step towards new applications for STIRAP, to prepare excitation regions or population patterns in space with large resolution.

Keywords: stimulated Raman adiabatic passage, electromagnetically induced transparency, subwavelength localization, adiabatic passage, rare-earth ion-doped solid

1. Introduction
Quantum memories are an integral part of any large scale quantum communication structure, e.g., as part of repeaters or processors. In simple terms, such a quantum memory temporarily stores the information encoded in the quantum state of a qubit. Hence, the most important characteristics of this memory are its fidelity, storage efficiency, time, and capacity [1]. Optical implementations of quantum memories use photons as information carriers and often ensemble-based media, e.g., cold atomic gases, warm vapors or emitters in solid-state systems such as rare-earth ion-doped crystals [2]. The latter solid-state systems hold significant potential for realistic applications in quantum technologies, because they combine free-atom-like, spectrally narrow transitions [3] with the advantages of solids, i.e., absence of atomic motion, robust handling, scalability, and integrability [2].

There are already some impressive results on implementations of quantum memories in rare-earth ion-doped crystals, reaching, e.g., storage efficiencies up to 76% [4], storage times up to 53 min [5] and fidelities of 99.9% [6]. In order to increase the capacity of the memory, i.e., its ability to store more than one qubit simultaneously, most approaches so far rely on temporal [7, 8] or spectral [9] multiplexing. While storage of up to 1060 modes was demonstrated [8],...
these techniques are inherently limited by the bandwidth of the memory protocol and medium. An alternative approach utilizes spatial multiplexing. Our team already implemented such multiplexing by exploiting the phase matching condition of the memory protocol to store information in overlapping interference patterns in the crystal [9]. However, the available angular resolution strongly limits this approach and it may suffer from cross-talk between information channels. A much simpler approach to spatial multiplexing relies on selective addressing of individual storage volumes in an extended medium [10]. The larger the medium and the smaller the optically addressable volume, the larger the spatial storage capacity of the memory. In this case, the diffraction limit for focused laser beams is expected to limit the storage capacity. Hence, we require methods to optically address small sections of a memory, potentially with extensions of the excitation area below the diameter of the driving laser beam, even if focused down to the diffraction limit. In the following, we will deal with novel techniques to overcome the diffraction limit in optical excitation.

We note, that there are a multitude of other applications beyond the storage capacity in an optical quantum memory, which benefit from spatial confinement of optical excitations below the diffraction limit. Examples from quantum technology are selective addressing of single emitters from a large ensemble as isolated qubits [11–13], the generation of narrow waveguide-like excitation structures in crystals to enhance light–matter interaction for quantum information processing [14], patterning of Bose–Einstein condensates [15, 16], or single site addressing below the diffraction limit in a Paul ion trap or optical lattice [17–19]. So far there are no experimental implementations of STIRAP driven localization at all, nor does not permit population transfer to a metastable state. Nevertheless, there are no implementations of EIT for localized excitations in solids yet. We also note, that all implementations so far applied a standing wave geometry, which leads to an extended one-dimensional chain of excitations. In our experiment we use a STED-like geometry which yields a single localization spot in two dimensions [16, 19].

More recently, Mompart and co-workers proposed to apply stimulated Raman adiabatic passage (STIRAP) [38, 43] for subwavelength localization [15]. As an important finding from the theoretical treatment, the nonlinearity in STIRAP is much steeper compared to EIT. Thus, the spatial resolution increases much faster with increasing laser intensities for STIRAP than for EIT. In other words, at a given laser intensity, the localized excitations by STIRAP are much tighter spatially confined compared to EIT-based approaches [15, 16, 19]. So far there are no experimental implementations of STIRAP-driven localization at all, nor matter in which medium.

In the following work, we present a convincing experimental demonstration and thorough systematic study of localized excitations by STIRAP in a rare-earth ion-doped crystal, and compare the results to EIT as well as numerical simulations. The proof-of-principle experiment operates at dimensions above the diffraction limit. Nevertheless, the experimental data fully confirm the theoretical predictions and pave the way for further investigations towards subwavelength resolution.
2. Basic theory

We review now the basic theory of localized excitations, driven by EIT [23, 40] or STIRAP [15, 19]. The adiabatic passage processes rely on coherent interaction in a Λ-type three-level system with two metastable ground states, coupled to a single excited state by two laser pulses (see figure 1(a)). The pump drives transition $|1\rangle \leftrightarrow |2\rangle$ with Rabi frequency $\Omega_P$, the Stokes drives transition $|3\rangle \leftrightarrow |2\rangle$ with Rabi frequency $\Omega_S$. We note, that for consistency, throughout the paper we use the typical STIRAP nomenclature ‘pump’ and ‘Stokes’ also for EIT, where the pulses are otherwise usually termed ‘probe’ and ‘control’. The dressed eigenstates of this coupled light–matter system include a so-called dark state given by [39]

$$|D(r)\rangle = \frac{\Omega_S(r)|1\rangle - \Omega_P(r)|3\rangle}{\sqrt{\Omega_P^2(r) + \Omega_S^2(r)}}. \quad (1)$$

Since the dark state does not contain any contribution of the excited state $|2\rangle$, an atom in this state is essentially trapped therein, i.e., it can no longer be optically excited. This phenomenon is usually referred to as coherent population trapping.

Let us first consider localization by EIT. We assume that the system is in the dark state (and will later discuss how to transfer it into this state). In this case, the population distribution of the bare states $|1\rangle$ and $|3\rangle$ depends only upon the ratio $\Omega_P(r)/\Omega_S(r)$ and, hence, it varies across the laser intensity beam profiles. As an instructive example, let us assume a beam geometry similar to STED, i.e., a Laguerre–Gaussian ‘donut’ mode for the pump beam profile

$$\Omega_P(r) = \Omega_{P0} \frac{r}{w_P} e^{-r^2/w_P^2} \quad (2)$$

and a Gaussian mode for the Stokes beam profile

$$\Omega_S(r) = \Omega_{S0} e^{-r^2/w_S^2} \quad (3)$$

Here, $r$ is the radial coordinate, $\Omega_{P0}$ and $\Omega_{S0}$ are the peak coupling strengths, and $w_i$, with $i = P, S$, are the beam waists. Close to the node of the pump beam we have $\Omega_P \ll \Omega_S$. Thus, we get $|D\rangle = |1\rangle$ at the node, whereas everywhere else (provided that $\Omega_{P0} > \Omega_{S0}$) we have $|D\rangle \approx |3\rangle$. Hence, as a consequence of the spatial variation in $\Omega_P(r)/\Omega_S(r)$, only in a spatially tightly confined region around the intensity node of the pump beam the system remains in state $|1\rangle$, i.e., the population $P_1$ defined as the density matrix element $\rho_{11}$ has a sharp peak at the pump intensity node (see figure 1(b)). The population in state $|3\rangle$ shows the opposite behaviour, i.e., a sharp dip at the center of the donut pump profile (not depicted in figure 1(b)). We note, that it would easily be possible to exchange the occurrence of a peak or a dip in the spatial population patterns of the two states $|1\rangle$ and $|3\rangle$, by preparing the system initially in the other ground state and exchanging the beam profiles of pump and Stokes. In any case, the localization gets tighter when $\Omega_{P0}$ increases, as the condition $\Omega_P > \Omega_S$ is fulfilled closer to the center of the node. A simple calculation (see appendix A) shows that the full width at half maximum (FWHM) of the population distribution is

$$\Delta r = \frac{2w_P}{\sqrt{R}} \quad (4)$$

where we define $R = \Omega_{P0}^2/\Omega_{S0}^2$. This clearly shows, as expected, that the localization improves for increasing $R$, i.e., increasing pump Rabi frequency—which is proportional to the electric field of the driving radiation.

There are several ways to transfer the system to the dark state. Let us assume, that initially the population is in state $|1\rangle$. The initial proposal suggested coincident pump and Stokes pulses with equal temporal pulse intensity profiles to optically pump the system to the dark state [40]. This requires however, that the pulses are much longer than the lifetime $T_1$ of the excited state $|2\rangle$. Otherwise, the localized population pattern in space broadens [44] and diabatic coupling causes a complicated, oscillatory behavior in the wings [42]. Furthermore, the transfer involving decay is incoherent and not appropriate for applications in, e.g., quantum technology. Therefore, following work on EIT-based localization [23, 31, 41, 42] used a counter-intuitive sequence of shaped pulses. This already resembled fractional STIRAP [43]. Here, the Stokes pulse precedes the pump pulse such that initially $|D\rangle = |1\rangle$. If the pump beam then turns on sufficiently slow and temporally overlapping with the Stokes pulse, the system adiabatically follows the dark state into the spatially varying superposition of $|1\rangle$ and $|3\rangle$. Finally, the pulses ends simultaneously to maintain a constant ratio $\Omega_P(r)/\Omega_S(r)$ which ensures that the system remains in the dark state. Compared to the initial proposal [40], this approach maintains coherence. However, the achieved spatial confinement of the excitation process remains just the same. Moreover, the protocol is very sensitive to variations of the shape in the falling edges of the pulses.

In more recent theory work, Mompart and co-workers proposed to apply the full, standard STIRAP process for subwavelength localization [15]. This approach requires appropriately delayed pump and Stokes pulses only, without the need for specific pulse shapes, e.g., in the falling edges. For STIRAP, the Stokes pulse precedes the pump pulse. At early time, when $\Omega_P < \Omega_S$, the system is in the dark state $|D\rangle = |1\rangle$. At late
time, when $\Omega_s < \Omega_p$, the dark state has evolved into the target state as $|D\rangle = |3\rangle$. This is the essence of complete, adiabatic population transfer from state $|1\rangle$ to state $|3\rangle$ by STIRAP. The process is robust, i.e., the population dynamics do not change when the experimental parameters fluctuate—provided some limits are kept in view. If we apply the geometry of a donut pump and a Gaussian Stokes beam for STIRAP, no population will be transferred in a tight region around the node of the pump field, whereas the robustness of STIRAP ensures almost perfect transfer everywhere else. The larger the peak intensity in the beams, the tighter the region where population remains in state $|1\rangle$. Efficient STIRAP requires us to fulfill the global adiabaticity condition [37]

$$\Omega_s^2 + \Omega_p^2 \geq \left(\frac{A}{T}\right)^2,$$  \hspace{1cm} (5)

where $T$ is the delay between the pulses and $A$ is a minimal pulse area (i.e., product of Rabi frequency and pulse duration) necessary for efficient transfer. Typically, STIRAP demands $A \gg 10$ [37]. In the terminology of incoherent excitation, the large pulse area simply corresponds to saturation of a transition. We follow the ansatz by Viscor and coworkers [19] (see appendix A) to find the diameter (FWHM) of the region around the pump node, where the atoms are left in state $|1\rangle$:

$$\Delta r = \sqrt{\frac{2}{w_p} \frac{A^2}{R} \frac{1}{\left(\frac{A}{T\Omega_p}\right)^2 - 1}}.$$ \hspace{1cm} (6)

Analysis of equations (4) and (6) reveals that STIRAP reaches much stronger spatial confinement of population patterns and converges much faster with increasing laser intensity towards tight localization compared to EIT (see figure 1(b)) and converges much faster with increasing laser intensity towards tight localization compared to EIT (see figure 1(b)) and converges much faster with increasing laser intensity towards tight localization compared to EIT (see figure 1(b)) and converges much faster with increasing laser intensity towards tight localization compared to EIT (see figure 1(b)) and converges much faster with increasing laser intensity towards tight localization compared to EIT (see figure 1(b)) and converges much faster with increasing laser intensity towards tight localization compared to EIT (see figure 1(b)). Moreover, the pulse sequence of STIRAP is easy to implement and robust with regard also to fluctuations in the pulse shape or other experimental parameters. The process maintains coherence and produces no recoil.

3. Experimental setup

We implement our experiments on localized excitations by adiabatic passage in a Pr$^{3+}$:Y$_2$SiO$_5$ (from now on simply termed Pr:YSO) crystal, among hyperfine states of the optical transition $^3\!H_4 \leftrightarrow ^1\!D_2$, at a center wavelength of 605.98 nm. We choose the hyperfine states $|1\rangle = ^3\!H_4|m = +3/2\rangle$, $|3\rangle = ^3\!H_4|m = +1/2\rangle$, and $|2\rangle = ^1\!D_2|m = +1/2\rangle$ for our experiments due to their corresponding favorable transition moments but note that in principle any of the hyperfine states can by utilized for localization. The excited state population lifetime in Pr:YSO is $T_1 = 164 \mu$s, whereas the ground state lifetime is $T_1 = 100 \mu$s. The latter is an attractive feature, as the population patterns in space driven by STIRAP or EIT live very long. The decoherence time $T_2 = 500 \mu$s among the ground states sets an upper limit for the interaction time with the driving light fields, i.e., the maximal pulse duration and pulse delay.

The optical transition in Pr:YSO is inhomogeneously broadened to several GHz, while the hyperfine state separation is only on the order of $\sim 10$ MHz. Hence, a single frequency laser couples all possible transitions in ions from different frequency ensembles within the inhomogeneous linewidth. Thus, we use an optical pumping sequence to prepare the required level scheme and population distribution. This preparation sequence starts by burning a spectral pit that contains both the pump and Stokes transition, followed by a repumping pulse to create an antihole on the pump transition with a residual inhomogeneous broadening below 100 kHz. Finally, a cleaning pulse removes unwanted population from state $|3\rangle$ to create the desired $\Lambda$ system with the population initially in state $|1\rangle$ (see figure 1(a)). For more details on the preparation sequence see [45, 46].

Figure 2(a) depicts our experimental setup around a Pr:YSO crystal (length 1 mm, dopant concentration 0.05%) which we cool to temperatures below 4 K in a continuous flow cryostat (ST-100, Janis Research Co.). We derive all laser beams for the experiment from an optical parametric oscillator system (based on Argos Model 2400 SF-15, Lockheed Martin Aculight) [47]. It involves a PPLN crystal with separate sections for optical parametric oscillation and sum frequency generation placed in a bow-tie resonator. The two nonlinear optical processes convert the near infrared pump radiation from an amplified fiber laser into the visible regime around 606 nm. The system provides up to 400 mW of optical power at the experiment. We reduce its linewidth to well below 100 kHz (FWHM) using a Pound–Drever–Hall stabilization unit with a reference cavity. Each beam line in the experiment can be controlled in
intensity and frequency using acousto-optic modulators, to generate preparation, pump, Stokes, and probe pulses with appropriate temporal pulse shapes, durations and timings. The pump beam with an initially Gaussian intensity distribution in space passes a spiral phase plate with charge 1 (V-593-20-1, Vortex Photonics) which imprints an azimuthally increasing phase onto the beam. This converts the pump beam mode to a Laguerre–Gaussian ‘donut’-like intensity profile. We mildly focus the pump beam into the crystal with lens L1 (focal length 200 mm), yielding a beam waist of \( w_p = 100 \mu m \). The Stokes beam (which we also use for the optical preparation of the medium) counterpropagates to the pump beam with a small angle of about 2° in between. We collimate the Stokes beam into the crystal with lenses L2 (focal length 150 mm) and L3 (focal length 60 mm), yielding a beam waist of \( w_S = 250 \mu m \) at the position of the crystal. This large size compared to the pump beam ensures less modulation of the Stokes Rabi frequency and a homogeneous optical preparation of the medium across the interaction region.

To determine the spatially-varying population distribution after adiabatic passage, we measure the transmission across a probe laser beam profile, when we tune the probe laser frequency in the range of the pump and Stokes transitions. The probe beam copropagates with the pump beam, but is temporally well separated by roughly 7 ms with regard to the STIRAP pulses. The delay is much longer than the lifetime of the excited state \( |3\rangle \) which allows for residual phase oscillations. Hence, there will be no residual population in state \( |2\rangle \) when the probe pulse interacts with the medium, i.e., all population will be in the ground states \( |1\rangle \) and \( |3\rangle \). The probe beam has a beam waist of 290 \( \mu m \) in the crystal, i.e., much larger than the pump profile, in order to cover the full interaction region. We image the probe beam profile onto a CCD camera (Prosilica GC1290, Allied Vision) using a simple imaging system consisting of lenses L3 and L4 (focal length 300 mm).

We determined the magnification and resolution of the imaging system using a USAF-1951 target mask placed instead of the crystal in the beam lines. The measured resolution of 3 \( \mu m \) is only slightly larger than the diffraction limit given by the numerical aperture \( NA = 0.23 \) of the system. The magnification of 4.68 fits with the ratio of the focal lengths of lenses L2 and L3. A mechanical shutter prevents saturation of the camera caused by the pump beam or back reflections of the Stokes beam.

From the probe beam intensity profile at the CCD camera, we determine the probe transmission \( T_1 \) at the pump transition \( |1\rangle \rightarrow |2\rangle \) and \( T_3 \) at the Stokes transition \( |3\rangle \rightarrow |2\rangle \). We compare these values to the maximal transmission \( T_0 \) through the optical setup, measured during the preparation sequence, when the crystal should be fully transparent. After subtracting a background measurement, this allows us to determine the spatial distribution of the optical depths \( OD = \ln(T/T_0) \) at the pump and Stokes transition and, hence, the populations in the states \( |1\rangle \) and \( |3\rangle \). We note, that in principle also one transition would be sufficient to determine the population distribution in the initial and target state. However, measuring at both the pump and Stokes transition yields a larger signal-to-noise ratio.

Figure 2(b) summarizes the time sequence of our experiment: (i) preparation pulse sequence using optical pumping to provide full transparency for both Stokes and pump transition. (ii) Transmission measurement of a probe pulse as reference. (iii) Preparation pulse sequence using optical pumping to provide a \( \Lambda \)-system. (iv) STIRAP or EIT pulse sequence to drive localized excitations. (v) Transmission measurement of a probe pulse to determine population patterns generated by STIRAP or EIT. We repeat the sequence (i)–(v) and average the resulting images to reduce noise.

In order to compare the results of our measurements to a numerical simulation, we require exact values of pump and Stokes Rabi frequencies, as they vary across the laser beam profiles at the position of the crystal. It would not be very accurate to calculate the Rabi frequencies from the laser intensity beam profiles, as uncertainties in the intensity and transition moments would add up. Hence, we decided to directly measure the Rabi frequencies by spatially-resolved observation of the probe laser beam profile, when we tune the probe laser frequency to a point in time corresponding to the STIRAP/EIT. We repeat the sequence (i)–(v) and average the resulting images to reduce noise.

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4. Experimental results

4.1. Localized excitation by STIRAP

We discuss now our experiments on localization driven by STIRAP. We apply the time sequence discussed above and presented in figure 2(b). We choose pulses with a Gaussian intensity profile in time with a FWHM of \( \tau_p = \tau_S = 25 \mu s \), i.e., well below the ground state decoherence time \( T_2 = 500 \mu s \). The pump follows the Stokes pulse with a time delay of \( T = 25 \mu s \). We found that the exact value of the time delay does not matter as long as it is roughly equal to the pulse duration and hence chose \( T = \tau_p = \tau_S \). For systematic measurements we keep the peak Stokes Rabi frequency at \( \Omega_{S0} = 2\pi \times 225 \text{ kHz} \) and vary the peak pump Rabi frequency \( \Omega_{P0} \) from 0 to \( 2\pi \times 2710 \text{ kHz} \) (limited by the available laser power). For each value of \( \Omega_{S0} \) (i.e., ratio \( R \) of the peak Rabi frequencies), we measure the population \( P_1 \), with a probe pulse of Gaussian intensity profile in time with a FWHM of 10 \( \mu s \) and a peak Rabi frequency of roughly \( 2\pi \times 8 \text{ kHz} \) which leads to only negligible change of the population distribution. We expose the CCD camera to radiation for 40 \( \mu s \), which ensures detection of the entire probe pulse.
Figure 3. STIRAP-driven localization in Pr:YSO. Variation of the population $P_1(x, y)$ vs coordinates $x$ and $y$ across the laser beam profiles. (Upper row) Experimental data for different values of $R$. Blue corresponds to zero population, red to $P_1(x, y) = 1$. The white, dashed line indicates the diameter of the pump beam, defined by the position of the ring at maximal pump intensity. (Middle row) Numerical simulations, with the experimentally determined parameters. (Lower row) Cuts through the experimental data (red line), simulation (green line) and the spatially filtered simulation (black, dashed line). The plots presented in this figure show a small subset of our available data for four selected values of $R$. The supplemental material (https://stacks.iop.org/B/55/154003/mmedia) shows a video including plots for 50 values of $R$.

The top row of figure 3 shows the experimentally measured population distributions after the STIRAP process for selected values of $R$. The bottom row shows cuts (indicated by red lines) through the central peak. Obviously, the remaining population in state $|1\rangle$, described by $P_1$, is localized in the center of the donut pump beam. Already at $R = 0.75$, the population is well confined below the pump beam diameter. This is already clear evidence for adiabatically-driven localization. The extension of the population region shrinks for increasing pump laser intensity (i.e., larger values of $R$), as expected. We also see, that for ratios $R \geq 4$ the background population essentially reduces to zero, confirming the almost perfect transfer by STIRAP as soon as the pump Rabi frequency is sufficiently large to fulfill the global adiabaticity condition (5).

However, it is also apparent that for $R \geq 25$, the extension of the population confinement seems to remain approximately constant at $\Delta r \approx 15 \mu m$, while only its amplitude decreases further. This is due to the extension of the crystal in propagation direction and the limited depth of field in our imaging system. While our imaging system has a diffraction limited resolution of about $3 \mu m$ at its focus, the resolution for an object $0.5 \text{ mm}$ (i.e., half the crystal thickness) outside the focus is only about $10 \mu m$—which we confirmed by an optical measurement. Thus, even larger structures outside the focus are not well imaged and smear out. When we image the population distribution in the crystal, our imaging system averages this distribution in propagation direction. Therefore, the population distribution is washed out due to lower resolution at the edges of the crystal. We also note, that the maximal population in the center of the pump profile slightly deviates from the maximal value of one, as there is some residual intensity $>0$ in the pump node.

We compare the results of our measurement with a straightforward numerical simulation based on a density matrix calculation of the $\Lambda$-system in Pr:YSO, interacting with pump and Stokes pulses [39]. The calculation also takes decay of the excited state $|2\rangle$ and the limited decoherence time among the ground states $|1\rangle$ and $|3\rangle$ in Pr:YSO into account, as well as inhomogeneous broadenings of the optical and hyperfine transitions in the rare-earth ion-doped solid. In the calculation we use a fit to the spatial variation of pump and Stokes Rabi frequencies, as experimentally determined from measurements of Rabi oscillations (see above) and set all other pulse parameters to be the same as in the experiment. To reduce the required calculation time, we assume rotational symmetry in the beam profiles. The middle row of figure 3 shows the results of the simulation. The green lines in the bottom row indicate cuts through the central peak in the two-dimensional plots of the simulations. For low values of $R$ we see a good agreement between the simulations and the experimental data, with some offset in the wings. We suspect that this is caused by a small mismatch between the simulation parameters and the experiment. The sensitivity of STIRAP at low pump Rabi frequencies, e.g., as it is here the case in the wings of the beam profile, then causes the small visible deviation. At larger values of $R$ the simulation deviates from the experiment—as expected, when our imaging system washes the spatial confinements of narrow population distributions out. We mimic the latter imaging effect in our simulation by applying a simple two dimensional Gaussian filter with a FWHM of $12 \mu m$ and show the resulting profiles as black, dashed lines in the bottom row of figure 3. Now we see a good agreement with the experimental data (red lines), especially for large values of $R$. 
We analyze the experimental data now in more detail, in order to obtain some more information on the localization process also for large values of $R$, even if the imaging system washes out the effect of stronger spatial confinements. In figure 4 we plot the width (FWHM) of the region with population confined by STIRAP versus $R$ (red triangles). The plot also shows results from our simulation (green squares) and the analytic treatment (6) (black line) for which we used the experimentally determined parameters and $A = 38$. As discussed before, we see that the extension of the region with localized population is limited to $15 \mu$m and does not match the expectation. This is due to the limitations of the imaging system.

Let us draw our attention again to the two-dimensional plots in the top row of figure 3: we see, that for large values of $R$ the region with localized population does not shrink any further (as we already discussed), but it becomes fainter, i.e., the maximal signal decreases. We also see this in the cuts in the bottom row of figure 3 where the amplitude decreases for larger values of $R$. This can be explained by the fact, that the stronger STIRAP spatially confines the population, the fewer quantum systems are selected by the process and contribute to the signal. Hence, the integral of the signal is proportional to the number of quantum systems, spatially selected by STIRAP. This holds true, irrespective of the spatial resolution of the imaging system, i.e., if the measured width is limited by the imaging system, the amplitude has to decrease. This gives us a handle to infer the true width of the localized population also from resolution-limited experimental data. To do so, we rescale the fits to the experimental data (e.g., the red lines in the plots in the bottom row of figure 3), such that they reach a peak height of one, while reducing their width such that the integral remains the same. We show the resulting, recalibrated widths of the localized population as blue circles in figure 4. They are in better agreement now with the numerical simulation and analytic treatment. We assume that the remaining discrepancy is due to the reduced peak population caused by residual intensity in the center of the pump profile. We do not account for this behavior in our simple rescaling procedure which leads to an overcorrection of the data. Nevertheless, our experiment confirms the theoretical proposal of STIRAP-based localization and its analytic treatment (6). Under our experimental conditions with still very large beam diameter and small Rabi frequencies, STIRAP reaches a localization of at least $\Delta r \approx 3 \mu$m, i.e., very much below the pump beam diameter. Extrapolating from these results, already for still moderate focusing to a donut size of $w_p = 15 \mu$m, yielding (at the same pump pulse energy as in our present experiment) a pump Rabi frequency of $\Omega_{R0} = 2\pi \times 18$ MHz, we would confine the population to $\Delta r \lesssim 100$ nm, i.e., well below the diffraction limit.

4.2. Comparison with localized excitation by EIT

We proceed now to localization by EIT in order to compare the results with STIRAP-driven localization. We keep all experimental parameters as in the STIRAP measurements, except for the temporal shape of the Stokes pulse. The latter consists of a Gaussian rising and falling edge with a rise/fall time of $25 \mu$s (FWHM), and a plateau of constant intensity with a duration of $30 \mu$s in between. We choose the pulse delay such, that the falling edges of Stokes and pump coincide, as required for EIT-driven localization [23].

The top row of figure 5 shows the experimentally measured population distributions after the EIT process for selected values of $R$. The bottom row shows cuts (indicated by red lines) through the central peak. Clearly, population is localized in the center of the pump beam, with an extension well below the beam diameter. Comparison to STIRAP (see figure 3) very obviously shows the superior performance of STIRAP. For all values of $R$, STIRAP yields a much tighter localization than EIT. Furthermore, we see that STIRAP reaches full transfer efficiency in the outer parts of the pump profile already at $R = 4$, while for EIT even at the largest value $R = 145$, roughly 10% of the population remains in state $|1\rangle$. This strongly reduces the fidelity of EIT-driven localization. Hence, STIRAP clearly outperforms EIT, as predicted by theory [15, 16, 19].

The numerical simulation (see middle row of figure 5 and corresponding cuts in the graphs of the bottom row) fully confirms our findings and conclusion.

Let us finally consider the convergence of EIT to stronger confinement of populations with increasing values of $R$. In figure 6 we plot the width of the region with localized populations (FWHM), determined from fits to the experimental data (red triangles), numerical simulation (green squares), and analytic treatment (4) (black line) vs the ratio $R$. We also apply the recalibration method, as presented for STIRAP, to the experimental EIT data in order to get information on the true width of the spatially confined population (see blue circles). Note that due to the larger excitation regions in EIT-based localization, the limited resolution of the imaging setup and thus the recalibration has a much smaller effect here compared to the STIRAP data. We find very good agreement, in particular for the recalibrated data points compared to the numerical simulation. The slight, systematic deviation compared to the analytic treatment is due to inhomogeneous broadening, which we neglected in the analytic solution. For EIT we find
Figure 5. EIT-driven localization in Pr:YSO. Variation of the population $P_1(x, y)$ vs coordinates $x$ and $y$ across the laser beam profiles. (Upper row) Experimental data for different values of $R$. Blue corresponds to zero population, red to $P_1(x, y) = 1$. The white, dashed line indicates the diameter of the pump beam, defined by the position of the ring at maximal pump intensity. (Middle row) Numerical simulations, with the experimentally determined parameters. (Lower row) Cuts through the experimental data (red line), simulation (green line) and the spatially filtered simulation (black, dashed line).

Figure 6. Width (FWHM) of the population distribution $P_1(x, y)$ after localization by EIT vs $R$. Comparison of the width calculated from (4) (black line), numerical simulation (green squares), experimental data (red triangles) and recalibrated experimental data taking limited imaging resolution into account (blue circles). Note the different scale of the vertical axis compared to figure 4.

The strongest localization at $\Delta r \approx 20 \mu m$, i.e., a factor of 6.5 larger compared to STIRAP, at equal experimental parameters. From theory we calculate, that EIT would require more than 25 times larger pulse energy to match with STIRAP. Again, this reveals the superior performance of STIRAP-driven localized excitation compared to EIT.

5. Summary

We experimentally demonstrated localization of atomic populations by STIRAP and EIT in Pr:YSO. To the best of our knowledge, this represents the first implementation of EIT-driven localized excitations in a solid medium and the first implementation of the STIRAP-based approach at all. We apply a STED-like geometry with a Gaussian-shaped Stokes and a donut-shaped pump beam to confine population by STIRAP or EIT in the zero-intensity center of the pump beam profile. We determine the obtained population distribution across the laser beam profiles by spatially resolved absorption spectroscopy with a probe laser. In particular, our experiments confirm, that adiabatic passage confines atomic populations to spatial extensions well below the diameter of the driving laser beams. If we apply a pump beam with a beam waist of $w_P = 100 \mu m$ and peak Rabi frequencies of $\Omega_P = 2\pi \times 2710$ kHz, we get a spatial confinement of populations to $\Delta r \approx 3 \mu m$ for STIRAP and $\Delta r \approx 20 \mu m$ for EIT. We confirmed, that the localization improves with increasing laser intensity. Moreover, our data demonstrate that STIRAP converges to smaller excitation regions must faster compared to EIT when increasing the laser intensity. The latter would require more than an order of magnitude larger laser pulse energy to match with the performance of STIRAP. Hence, STIRAP-driven localization clearly outperforms EIT, as it was predicted by theory [15, 16, 19]. The experimental data agree with numerical simulations and the analytic treatment [19]. Residual deviations are due to slight parameter mismatches but mostly due to the limited resolution of our imaging setup. We note, that at our experimental conditions and due to the resolution limit of our imaging system, we still operated well above the diffraction limit. Nevertheless, the experiment permits extrapolation towards obtaining localized excitations in the sub-diffraction regime. With still moderate beam diameters and Rabi frequencies, spatial confinement of populations towards extensions $\leq 100$ nm in Pr:YSO is easily in reach. The results will pave the way for a novel application of STIRAP, to prepare population patterns or confine optical excitations in a medium with large spatial
resolution. This will be of relevance to quantum information technology and well beyond.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Appendix A. Derivation of the width of the population localized by EIT and STIRAP

The derivations in this appendix follow the approach by Viscor and coworkers [19].

In the case of EIT, the FWHM of the localized population is equal to this threshold to obtain the adiabaticity condition (5) to obtain the derivations in this appendix follow the approach by Viscor and coworkers [19].

(1) and using the beam profiles (2) and (3) it is easy to see that this is the case for

\[ \Omega P_{\text{FWHM}} = \frac{\Omega \rho_0}{w_p} \left( r_0 e^{-r_0^2/w_p^2} + \Omega_2 e^{-r_0^2/w_2^2} \right) \geq \left( \frac{A}{T} \right)^2. \]  \hspace{1cm} (A4)

Following the approach by Viscor and coworkers [19] we expand this equation to second order and use the equality in (A4) to define a spatial threshold \( r_{\text{th}} \) for successful STIRAP:

Finally, we solve for \( r_{\text{th}} \) and assume that the FWHM of the localized population is equal to this threshold to obtain

\[ \Delta r = r_{\text{th}} = w_p \sqrt{\frac{\Omega_2^2 r_0^2}{w_p^2} + \Omega_2^2 \left( 1 - \frac{2 r_0^2}{w_2^2} \right)} = \left( \frac{A}{T} \right)^2. \]  \hspace{1cm} (A5)

\[ \Delta r = r_{\text{th}} = w_p \frac{\sqrt{\frac{A^2}{\Omega_2^2} - 1}}{R - \frac{2 w_p}{w_2}}. \]  \hspace{1cm} (A6)

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