

Subwavelength spatial confinement of optical excitation by stimulated Raman adiabatic passage variants in a doped solid

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We experimentally demonstrate subwavelength spatial confinement of optical excitation using variants of stimulated Raman adiabatic passage (STIRAP) in a $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal. Building on conventional STIRAP, we implement two more advanced schemes, double STIRAP and tripod STIRAP, which enable enhanced spatial control. At the cost of reduced coherence, double STIRAP provides an almost twofold improvement in resolution compared with conventional STIRAP, which already enables localization nearly two orders of magnitude below the pump beam waist. To resolve such fine structures near the diffraction limit, we employ an autocorrelation-based measurement technique. With this setup, we demonstrate excitation confinement by conventional STIRAP down to 260 ± 40 nm, thus achieving subwavelength localization. As another powerful variant of adiabatic passage, we apply tripod STIRAP using an additional donut-shaped control beam that suppresses population transfer outside the beam center via coherent population trapping. With tripod STIRAP, we generate excitations as narrow as 350 ± 40 nm, a factor of 28 below the width of the driving control beam. We demonstrate the scalability of the method by preparing tightly packed grids with up to 41 excitation spots without crosstalk, making the method well-suited for quantum architectures. Our results establish STIRAP variants as robust, versatile tools for coherent, spatially selective optical excitation on a subwavelength scale, opening broad applications in quantum technologies and classical optics.

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I. INTRODUCTION

Precise spatial control of optical excitations is becoming increasingly important in quantum science, nanophotonics, and related fields, as the packing density of emitters increases toward ever larger numbers per unit area. Therefore, the ability to confine and manipulate optical excitations on subwavelength scales is crucial in quantum technologies (e.g., quantum computing) as well as in classical optics (e.g., high-resolution microscopy or lithography). Such capabilities are relevant in systems as diverse as atomic gases, trapped ions, nanoscale photonic interfaces, and ensembles of emitters in the solid state, and are essential for scalable architectures where spatial resolution directly determines performance.

Achieving control of spatially confined optical excitations while maintaining coherence remains a central challenge. Many powerful high-resolution techniques, such as stimulated emission depletion [1,2] or other nonlinear imaging schemes [3], achieve deep subwavelength confinement via incoherent and dissipative processes, but these unavoidably destroy phase information. While such techniques are suitable for

classical imaging, they are fundamentally incompatible with quantum technologies, which require preservation of coherence (e.g., for spatially selective addressing of qubits in ion traps or optical lattices [4–9]), improving the storage capacity of quantum memories [10–12], or spatially selective preparation and addressing of single emitters in large ensembles [13–15]. Hence, there is a need for approaches to confine optical excitations—potentially with subwavelength spatial resolution—while preserving coherence.

Adiabatic passage techniques open up a promising path toward such coherently driven spatial confinement [16,17]. By using the spatial variation of laser beam profiles and the intrinsically large nonlinearity of adiabatic population transfer, we can shape the dark state manifold to act as a highly selective spatial filter [18,19]. When Λ -type level configurations with long-lived ground states are utilized, the resulting spatially varying optical excitations may lead to long-lived atomic population patterns. Several theoretical proposals and first experimental demonstrations explored the idea of spatial confinement of optical excitations via adiabatic passage processes, including the well-known technique of stimulated Raman adiabatic passage (STIRAP) [12,19–21] and electromagnetically induced transparency (EIT) [5,22]. However, experimental progress remains limited, particularly for more advanced schemes such as tripod STIRAP [23], which have not yet been investigated experimentally.

In this work, we experimentally explore and compare several variants of STIRAP—namely, conventional STIRAP, double STIRAP, and tripod STIRAP [20]—to achieve localization of optical excitations in a rare-earth-ion-doped crystal.

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Such crystals are an established platform for quantum technologies. They offer long population and coherence lifetimes, along with the absence of atomic motion [10]. This permits the preparation of long-lived population patterns applicable to spatially multiplexed quantum memories [11], single-ion quantum computing [24], or high-fidelity laser stabilization [25]. As we aim for spatially confined optical excitation below the wavelength, we also implement an autocorrelation technique to probe excitation patterns with high spatial resolution. In particular, we analyze the performance of the STIRAP variants for localization in the subwavelength regime, their ability to preserve coherence, the improved confinement enabled by multiple applications of STIRAP, and the preparation of long-lived two-dimensional excitation patterns. As we demonstrate and discuss below, STIRAP and its variants prove to be versatile and powerful tools for coherently controlling optical excitations on subwavelength scales and even beyond the diffraction limit, with relevance for applications in quantum technologies as well as classical optics. A more comprehensive treatment of the theory, experimental procedure, and discussion presented here can be found in Ref. [26].

II. BASIC THEORY OF STIRAP-DRIVEN SPATIALLY CONFINED EXCITATIONS

We now introduce the basic principles for spatially confining (localizing) optical excitations, driven by conventional STIRAP, double (or multiple) STIRAP, and tripod STIRAP.

A. Localization by STIRAP and double STIRAP

STIRAP relies on coherent interaction in a Λ -type three-level system with two metastable ground states $|1\rangle$ and $|2\rangle$ and an excited state $|e\rangle$, which is subject to spontaneous decay. A pump beam drives the transition $|1\rangle \leftrightarrow |e\rangle$ with Rabi frequency Ω_P , and the Stokes beam drives the transition $|2\rangle \leftrightarrow |e\rangle$ with Rabi frequency Ω_S . We assume that the population is initially prepared in the ground state $|1\rangle$ and that the laser fields are tuned to resonance with the corresponding transitions [see Fig. 1(a)]. This configuration yields the following adiabatic eigenstate,

$$|\Psi(\mathbf{r})\rangle = \frac{\Omega_S(\mathbf{r})|1\rangle - \Omega_P(\mathbf{r})|2\rangle}{\sqrt{\Omega_P^2(\mathbf{r}) + \Omega_S^2(\mathbf{r})}}, \quad (1)$$

which is called the dark state [27] because it has no contribution from the excited state $|e\rangle$. To adiabatically transfer the population from state $|1\rangle$ to state $|2\rangle$, we apply a STIRAP sequence with the Stokes pulse preceding the pump pulse [see Fig. 3(b2), step (iv)]. In this case, at early times, the dark state $|\Psi\rangle$ aligns with the ground state $|1\rangle$. As we turn on the pump pulse while it still temporally overlaps with the Stokes pulse, the dark state continuously rotates toward the target ground state $|2\rangle$. The system remains in the eigenstate $|\Psi\rangle$ if the temporal variation of the Rabi frequencies $\Omega_{P,S}$ is slow compared with the evolution of the system governed by the Rabi frequencies $\Omega_{P,S}$. Thus, the temporal pulse shapes should be sufficiently smooth (e.g., Gaussian) to fulfill this criterion of “local” adiabaticity. Moreover, the pulse areas must be sufficiently large to fulfill the “global” adiabaticity

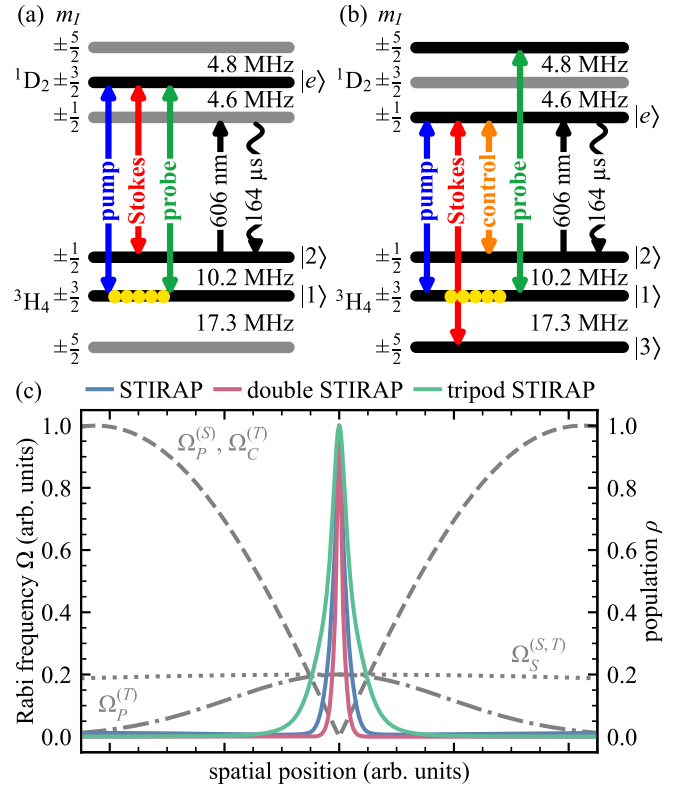


FIG. 1. Coupling schemes in Pr:YSO for (a) STIRAP or double STIRAP and (b) tripod STIRAP. (c) Localized population versus spatial coordinate across the laser beam profiles. For STIRAP and double STIRAP, we use a pump Rabi frequency $\Omega_P^{(S)}$ with a donut profile (gray dashed line) and a Stokes Rabi frequency $\Omega_S^{(S,T)}$ with a broad Gaussian profile (gray dotted line). For tripod STIRAP, we apply a control Rabi frequency $\Omega_C^{(T)}$ with the same donut profile (gray dashed line), while the pump $\Omega_P^{(T)}$ (gray dash-dotted line) is Gaussian and the Stokes $\Omega_S^{(S,T)}$ (gray dotted line) is identical to that used for STIRAP. The superscripts on the Rabi frequencies refer to the beam configurations used for STIRAP (S) or tripod STIRAP (T), with (S) and (T) denoting a beam shared by both.

criterion [17]

$$(\Omega_P^2 + \Omega_S^2)\tau^2 > A^2, \quad (2)$$

where τ is the delay between the pump and Stokes pulses, and A is typically a number with $A \geq 10$. If, for simplicity, we assume equal Rabi frequencies and a delay similar to the pulse duration, the latter condition translates to $\Omega\tau > 1$, which demands a large pulse area $\Omega\tau$ or, equivalently, strong driving and (in terms of incoherent excitation) saturation of the involved transitions. We note that efficient transfer is not achieved if only one beam is sufficiently strong to fulfill the global adiabaticity criterion while the other beam is weak.

We apply these findings to consider population transfer driven by STIRAP across spatially extended beam profiles. To this end, we assume a pump beam profile with a Laguerre-Gaussian TEM₀₁* “donut” mode

$$\Omega_P(\mathbf{r}) = \Omega_P \frac{\mathbf{r}}{w_P} e^{-\mathbf{r}^2/w_P^2} \quad (3)$$

and the Stokes beam with a simple Gaussian profile

$$\Omega_S(\mathbf{r}) = \Omega_S e^{-r^2/w_S^2}. \quad (4)$$

Here, $\mathbf{r}$ is the radial coordinate. Ω_i and w_i , with $i \in \{P, S\}$, denote the peak Rabi frequencies and beam waists of the pump and Stokes beams, respectively. We note that, in a typical experimental beam configuration, the Stokes beam diameter is chosen much larger than the pump beam diameter, i.e., $w_S \gg w_P$. This yields an almost constant Stokes Rabi frequency Ω_S across the donut-shaped pump beam. Intuitively, in this configuration, the population at the node of the pump profile ($r = 0$) is never excited because the pump field Ω_P vanishes there. Even close to the node ($r \approx 0$), population transfer remains small because the adiabaticity criterion is not satisfied. Hence, near the center of the pump beam, the system remains in state $|1\rangle$. Sufficiently far from the node, where the pump pulse area becomes large enough, we obtain highly efficient adiabatic population transfer to state $|2\rangle$. Between these two regimes, some diabatic transfer via the excited state $|e\rangle$ also occurs. In summary, we obtain a well-localized population peak remaining in state $|1\rangle$ at the center of the profile, while the population is transferred to state $|2\rangle$ everywhere else [see Fig. 1(c)]. This localization via STIRAP improves when the ratio

$$R = \frac{\Omega_P^2}{\Omega_S^2} \quad (5)$$

of the pulse energies, which are proportional to the squares of the peak Rabi frequencies Ω_i , increases. In fact, the size of the population distribution Δr in terms of full width at half maximum (FWHM) is given by [12]

$$\Delta r = w_P \sqrt{\frac{\left(\frac{A}{\tau \Omega_S}\right)^2 - 1}{R - 2 \frac{w_P^2}{w_S^2}}}. \quad (6)$$

Due to this pronounced nonlinearity with respect to the pulse Rabi frequency, the region in which the population remains in state $|1\rangle$ shrinks rapidly with increasing pulse energy to values far below the diameter of the driving laser beams, without being limited by diffraction.

In previous work, we demonstrated the ability of STIRAP to confine excitations below the diameter of the driving laser beams [12], but only for spatial extensions still far above the wavelength. In the work discussed below, we push STIRAP toward tighter confinement and also apply double STIRAP toward this goal. Double STIRAP is a straightforward extension: it entails applying STIRAP twice in a row, with a cleaning step in between, to avoid transferring the population from state $|2\rangle$ back to $|1\rangle$ via bright STIRAP [20]. The population distribution after two STIRAP processes is given by the square of the population distribution $\rho(\mathbf{r})$ after a single STIRAP process. This reduces the spatial width of the population peak in state $|1\rangle$ by a factor of 2 and hence improves spatial confinement by the same factor [see Fig. 1(c)]. In principle, the idea can be extended to an arbitrary number of subsequent STIRAP processes, as this simply multiplies the single-transfer profile again and again. Hence, if we apply STIRAP n times, the width of the spatially confined population decreases by a factor of n . Thus, localization significantly

improves with multiple applications of STIRAP. However, we note that, in our case, the cleaning step between transfers makes the process incoherent because we optically pump the population to another state. Irrespective of this additional requirement, multiple STIRAP confines atomic populations very efficiently in space, which is already relevant for applications. In the experiments discussed in the sections below, we demonstrate confinement of optical excitations by STIRAP and double STIRAP with spatial resolution reaching the subwavelength regime.

B. Localization with tripod STIRAP

As discussed above, STIRAP spatially localizes population in the initial state rather than confining optical excitation in a target state. Additionally, it suffers from diabatic transfer via the excited state in the wings of the beam profile. To overcome these issues, we now introduce tripod STIRAP [23] as an approach to spatially confine optical excitations. In addition to a Λ -type level scheme, the tripod scheme involves a third metastable ground state. The three ground states are coupled to the excited state by the pump, Stokes, and control beams. For an optimal choice of oscillator strengths on the relevant transitions in our specific medium Pr:YSO (see Table I), we assume the following couplings: the pump drives the transition $|1\rangle \leftrightarrow |e\rangle$, the Stokes field couples $|3\rangle \leftrightarrow |e\rangle$, and the control drives $|2\rangle \leftrightarrow |e\rangle$ (see Fig. 1(b)). We still apply pump and Stokes pulses in the usual STIRAP pulse order, with the Stokes pulse preceding the pump. The control pulse has a rectangular temporal shape, starts before the pump–Stokes STIRAP sequence, and shuts off immediately afterward [see Fig. 3(c2), step (iv)]. This tripod coupling scheme yields two degenerate dark states as adiabatic eigenstates:

$$|\Psi_1\rangle = -\cos \phi |1\rangle + \sin \phi |2\rangle, \quad (7)$$

$$|\Psi_2\rangle = \cos \theta \sin \phi |1\rangle + \cos \theta \cos \phi |2\rangle - \sin \theta |3\rangle, \quad (8)$$

with the mixing angles $\tan \phi = \Omega_P/\Omega_C$ and $\tan \theta = (\Omega_P^2 + \Omega_C^2)^{1/2}/\Omega_S$. From these equations, we see that, without the control beam, i.e., for $\Omega_C = 0$, we obtain $\phi = \pi/2$, and the state $|\Psi_2\rangle$ reduces to the dark state of conventional STIRAP:

$$|\Psi_2\rangle = \cos \theta |1\rangle - \sin \theta |3\rangle, \quad (9)$$

with $\tan \theta = \Omega_P/\Omega_S$, while the dark state $|\Psi_1\rangle$ reduces to $|2\rangle$. Hence, without the control pulse, we obtain high-fidelity STIRAP transfer from $|1\rangle$ to $|3\rangle$, provided that adiabaticity is maintained. We now consider the case $\Omega_C \neq 0$, in particular $\Omega_C \gg \Omega_P \approx \Omega_S$. This corresponds to a strong control pulse that acts as an envelope for the pump and Stokes fields at all times, which yields $\phi = 0$ and $\theta = \pi/2$. Therefore, the dark states reduce to the bare atomic eigenstates $|\Psi_1\rangle = |1\rangle$ and $|\Psi_2\rangle = |3\rangle$; that is, the population in $|1\rangle$ and $|3\rangle$ remains trapped. This phenomenon is known as coherent population trapping (CPT) [27]. Thus, the control beam enables gating of the high-fidelity STIRAP transfer: if the control beam is weak, STIRAP efficiently transfers population between metastable states, whereas for a strong control beam, STIRAP transfer is suppressed.

To apply this feature of tripod STIRAP to the localization of optical excitations, we choose pump and Stokes beams with

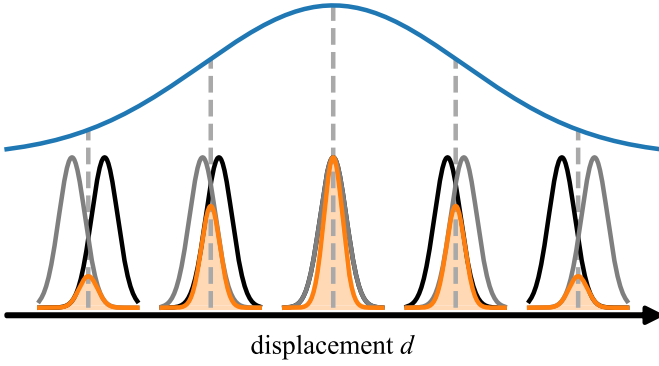


FIG. 2. Schematic illustration of an autocorrelation-based measurement. The black line depicts the spatial profile of the atomic population $\rho(r)$ after a first adiabatic transfer process. We then shift the laser beams in space (see main text) and drive the process a second time. The gray line depicts the population distribution $\rho(r-d)$ that would result from the second process. The resulting population profile (orange line) is given by the product $\rho(r)\rho(r-d)$ of the first (black line) and second (gray line) transfers. Integrating over the resulting profiles (orange shaded areas) for various displacements d yields the convolution $\int \rho(r)\rho(r-d)dr$ of the localized peak with itself, i.e., the autocorrelation function $\alpha(d)$ (blue line).

Gaussian profiles [see Eq. (4)] and a control beam with a donut-shaped profile [see Eq. (3)]. At the node of the donut, we have $\Omega_C \ll \Omega_P \approx \Omega_S$, i.e., STIRAP efficiently transfers population from $|1\rangle$ to $|3\rangle$. Outside the node, we approach the regime $\Omega_C \gg \Omega_P \approx \Omega_S$, i.e., STIRAP is suppressed and the population is trapped in $|1\rangle$ and $|3\rangle$ via CPT. Hence, efficient optical excitation is confined to the center of the donut-shaped control beam profile [see Fig. 1(c)]. Similar to conventional STIRAP, the extent of the excitation region shrinks rapidly with increasing laser intensity, i.e., the spatial resolution increases. In the intermediate regime, on the slopes of the control beam profile where $\Omega_C \approx \Omega_P \approx \Omega_S$, there is fractional transfer from $|1\rangle$ to $|3\rangle$, but no diabatic contribution from $|e\rangle$, as would be the case for conventional STIRAP. Hence, spatial confinement by tripod STIRAP fully preserves coherence.

An important advantage of tripod STIRAP is that it localizes the population in a target (excited) state rather than in the initial (ground) state. This allows us to prepare a tightly confined spot of optical excitation (or population transfer) in a medium and then shift the laser beams to prepare a second excitation spot nearby without affecting the first one. Thus, tripod STIRAP permits both the preparation and the selective addressing of tightly spaced excitation spots in a medium, i.e., the creation of two-dimensional atomic population patterns with a feature size (the excitation-spot width) much smaller than the dimensions of the laser beams. We demonstrate this feature of tripod STIRAP in the experiments discussed below.

III. MEASURING SPATIALLY CONFINED EXCITATIONS BY AUTOCORRELATION

As we aim to generate small excitation regions on sub-wavelength scales, we can no longer directly measure the resulting excitation patterns using simple, diffraction-limited

optical imaging, as in previous work [8,12,21]. Therefore, we implemented an autocorrelation-based measurement technique for STIRAP (similar to the approach used for EIT in Ref. [28]). Autocorrelation techniques are not limited by diffraction and thus enable the determination of excitation profiles at subwavelength dimensions. The concept is illustrated in Fig. 2. Suppose we first prepare a localized peak of atomic population $\rho(\mathbf{r})$ driven by STIRAP in our medium (see black line in Fig. 2). Afterward, we clean the population in state $|2\rangle$ by optical pumping to avoid transferring population back to $|1\rangle$ via bright STIRAP in the next step; that is, we ensure unidirectionality of the transfer. We then shift the beam profiles (gray line) by a small displacement $\mathbf{d}$ and drive the transfer process a second time, yielding the final population distribution (orange line):

$$\rho_{\text{res}}(\mathbf{r}, \mathbf{d}) = \rho(\mathbf{r})\rho(\mathbf{r} - \mathbf{d}), \quad (10)$$

with the resulting population profile ρ_{res} , the single-transfer profile ρ , the displacement $\mathbf{d}$, and the spatial coordinate $\mathbf{r}$. This procedure is repeated for various displacements $\mathbf{d}$, and the resulting $\rho_{\text{res}}(\mathbf{r}, \mathbf{d})$ is integrated over the whole image I :

$$\alpha(\mathbf{d}) = \iint_I \rho_{\text{res}}(\mathbf{r}, \mathbf{d}) d\mathbf{r} = \iint_I \rho(\mathbf{r})\rho(\mathbf{r} - \mathbf{d}) d\mathbf{r}, \quad (11)$$

which yields the autocorrelation function $\alpha(\mathbf{d})$. Assuming a spatially confined excitation profile, i.e., $\rho_{\text{res}}(\mathbf{r}, \mathbf{d}) = 0$ for $|\mathbf{r}| > R$, we can substitute the integration boundaries accordingly,

$$\alpha(x, y) = \int_{-R}^R \int_{-R}^R \rho(x', y') \rho(x - x', y - y') dx' dy'. \quad (12)$$

After performing a Fourier transform, we utilize the discrete convolution theorem [29] to get

$$\hat{\alpha}(m, n) = 4R^2 \hat{\rho}^2(m, n), \quad (13)$$

where the

$$\hat{\rho}(m, n) = \frac{1}{4R} \int_{-R}^R \int_{-R}^R e^{-2\pi i \frac{mx+ny}{2R}} \rho(x, y) dx dy \quad (14)$$

are the discrete Fourier coefficients, and $4R^2$ is a normalization factor. Using the convolution theorem from Eq. (13), we obtain an expression to retrieve the original excitation profile $\rho(\mathbf{r})$ from the autocorrelation function $\alpha(\mathbf{d})$:

$$\rho(\mathbf{r}) = \mathcal{F}^{-1} \left(\sqrt{\frac{\mathcal{F}(\alpha(\mathbf{d}))}{4R^2}} \right). \quad (15)$$

Thus, we obtain the original profile by measuring the autocorrelation function, performing a Fourier transform, normalizing, taking the square root, and applying an inverse Fourier transform. The spatial resolution of $\rho(\mathbf{r})$ is limited only by the sampling rate of the autocorrelation function. In our experiments, the limiting factor is the accuracy of the beam displacement $\mathbf{d}$, which is about 40 nm for our setup (see Sec. IV).

In the actual experiment, a few additional steps are required beyond applying Eq. (15). These include careful low-pass filtering of the data, interpolation, and extension of the autocorrelation function to decrease measurement time, improve resolution, and remove artifacts (see Appendix A). Moreover,

TABLE I. The relative oscillator strengths of all nine possible transitions between the hyperfine levels of the ground state $^3\text{H}_4$ and excited state $^1\text{D}_2$. The relevant transitions for STIRAP are highlighted in red and for tripod STIRAP in green. The values are taken from Ref. [30].

	m_l	$ e_1\rangle$ $\pm 1/2$	$ ^1\text{D}_2\rangle$	
			$ e_2\rangle$ $\pm 3/2$	$ e_3\rangle$ $\pm 5/2$
$ ^3\text{H}_4\rangle$	$ g_3\rangle$	$\pm 1/2$	0.55	0.38
	$ g_2\rangle$	$\pm 3/2$	0.4	0.6
	$ g_1\rangle$	$\pm 5/2$	0.05	0.93

autocorrelation measurements with tripod STIRAP require a slight modification of the algorithm because the population transfer is inevitably bidirectional; i.e., the second displaced tripod STIRAP sequence transfers population from the initial to the excited state and vice versa, which must be taken into account (see Appendix B).

IV. EXPERIMENTAL SETUP

We performed our experiments on the optical transition between the ground state $^3\text{H}_4$ and the excited state $^1\text{D}_2$ in a Pr:YSO crystal (dopant concentration 0.05%, length 1 mm). The transition wavelength is 605.98 nm. The ground and optically excited states exhibit hyperfine splitting, yielding a manifold of three levels each in the ground and excited states [see Figs. 1(a) and 1(b)] and nine possible optical transitions with different oscillator strengths (see Table I). The transitions are strongly inhomogeneously broadened to a spectral bandwidth of several GHz, which makes prior preparation by optical pumping necessary (see Refs. [30,31] for details on the preparation sequences). To implement the Λ -type level scheme for STIRAP and double STIRAP, we use the metastable ground states $|1\rangle = ^3\text{H}_4 |m_l = \pm 3/2\rangle$ and $|2\rangle = ^3\text{H}_4 |m_l = \pm 1/2\rangle$, together with transitions to the excited state $|e\rangle = ^1\text{D}_2 |m_l = \pm 3/2\rangle$. For the tripod STIRAP level scheme, we additionally utilize the third ground state $|3\rangle = ^3\text{H}_4 |m_l = \pm 5/2\rangle$ and transitions to the excited hyperfine state $|e\rangle = ^1\text{D}_2 |m_l = \pm 1/2\rangle$. The latter provides a better combination of oscillator strengths on the pump, Stokes, and control transitions (see Table I).

The coherence time among the metastable ground states is $T_2^g = 500 \mu\text{s}$, well above the typical total duration of the STIRAP pulse sequences (below $50 \mu\text{s}$). The population lifetime of the metastable ground states exceeds $T_1^g = 100 \text{ s}$, which permits the preparation of very long-lived atomic population patterns.

Figure 3(a) shows the experimental setup around the continuous-flow cryostat (ST-100, Janis Research Co.), which cools the Pr:YSO crystal to cryogenic temperatures below 4 K. The laser radiation for the experiments is provided by an amplified diode-laser system with an internal frequency-doubling unit (DLC TA-SHG pro 24509, Toptica Photonics), which delivers an output power of 800 mW with a frequency stability of about 20 kHz (FWHM). The laser beam is split into four beam paths by $\lambda/2$ plates and polarizing beam splitters. In these beam paths, we use acousto-optical

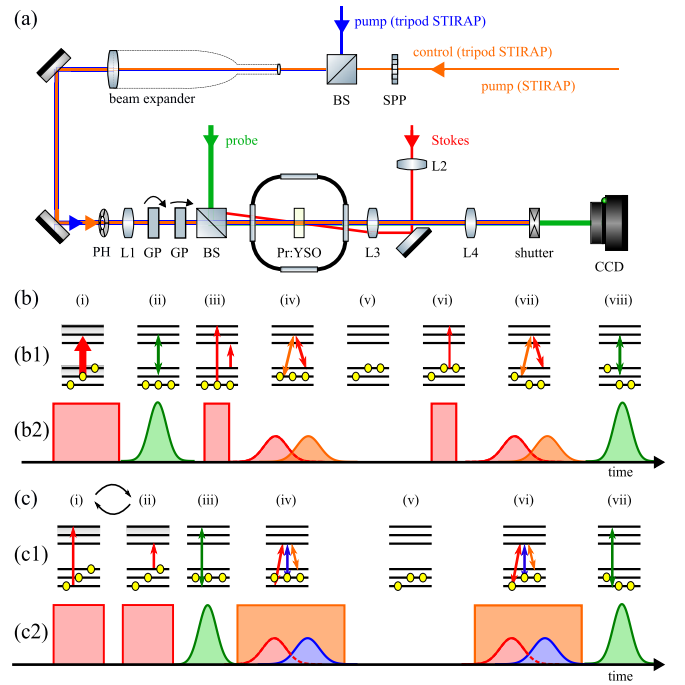


FIG. 3. (a) Experimental setup with pump, Stokes, control, and probe beams; lenses (L); spiral phase plate (SPP); beam splitters (BS); pinhole (PH); rotatable glass plates (GP); and a CCD camera. (b) Temporal sequence for STIRAP autocorrelation with (b1) laser pulses and (b2) the resulting population distribution. The sequence consists of (i) preparation of a spectral pit, (ii) probe absorption reference measurement, (iii) repumping and cleaning, (iv) first STIRAP sequence, (v) beam displacement, (vi) cleaning, (vii) second STIRAP sequence, and (viii) probe measurement. Steps (v)–(vii) are skipped for direct imaging of the population distribution. (c) Temporal sequence for tripod STIRAP autocorrelation with (c1) laser pulses and (c2) the resulting population distribution. The sequence consists of (i) preparation of a spectral pit around state $|g_1\rangle$, (ii) preparation of a spectral pit around state $|g_3\rangle$, (iii) probe absorption reference measurement, (iv) first tripod STIRAP sequence, (v) beam displacement, (vi) second tripod STIRAP sequence, and (vii) probe measurement. Steps (v) and (vi) are omitted for direct imaging of the population distribution. The colors used in (b2) and (c2) correspond to the beam paths in panel (a).

modulators in a double-pass configuration to generate probe, pump, Stokes, and (for tripod STIRAP) control pulses, which we can precisely control in intensity and frequency. For frequency modulation, we use direct digital synthesis (DDS) drivers (probe and pump: AODS 20160–1, Crystal Technology; Stokes: DDSPA-B8b23b-0, AA OptoElectronic; control: Model 409b, Novatech Instruments). The temporal shapes are controlled by arbitrary waveform generators for the probe and pump beams (33220A, Agilent). For the Stokes and control beams, the temporal shapes are set directly by the DDS drivers.

Initially, all laser beams have Gaussian profiles. The pump beam for STIRAP and double STIRAP must be modified to a donut profile. It therefore first passes a spatial filter to ensure optimal beam quality and low ellipticity. Afterward, a spiral phase plate with charge 1 (V-593–20–1, Vortex Photonics) imprints an azimuthally increasing phase onto the beam. This

results in a Laguerre-Gaussian TEM01* donut-shaped mode, as required for localization by STIRAP. The beam passes an adjustable beam expander (ZBE4A, Thorlabs), yielding a beam waist down to 10 μm after being focused by lens L1 (focal length 200 mm). After the lens, we use two glass plates of thickness 1 mm (WG11010-A, Thorlabs), with their tilt angles controlled by stepper motors. They are used to shift the beam in the horizontal and vertical directions via refraction for autocorrelation measurements and, as we demonstrate with tripod STIRAP below, to prepare a two-dimensional grid of atomic populations. A pinhole placed directly before the focusing lens L1 blocks up to 70% of the pump beam to ensure a clean donut-shaped profile in the crystal. Without the pinhole, the donut beam might be asymmetric, resulting in a nonzero intensity at the center of the beam after focusing. We note that a clean TEM01* donut profile is obtained only in the vicinity of the focal plane. Beyond this range—in particular, about 0.5 mm behind the 1 mm crystal—the transverse intensity distribution evolves into a Bessel-like profile as a consequence of diffraction at the pinhole.

For the tripod STIRAP experiments, another Gaussian beam is introduced to serve as the pump. This beam is overlapped with the donut-shaped beam and focused to a waist of 4 μm to ensure full overlap.

A probe beam, which is also overlapped with the pump and control beams via another beam splitter (BS043, Thorlabs) in front of the cryostat, measures the absorption by the created two-dimensional excitation patterns. At the position of the crystal, it has a waist of $w_{\text{pr}} = 180 \mu\text{m}$. Afterward, the probe is imaged onto a CCD camera (Prosilica GC1290, Allied Vision) via lenses L3 (focal length 60 mm) and L4 (focal length 300 mm) with a magnification of about 4.97, matching the ratio of focal lengths. A mechanical shutter [32] directly in front of the camera ensures that only the probe pulse passes to avoid overexposure. We typically average over multiple measurement cycles to increase the signal-to-noise ratio in the images. We apply Beer's law to obtain the population distribution from the images, compared with a reference image at full transmission obtained during preparation.

We characterized the imaging system with a USAF-1951 target at the position of the crystal. Accounting for the additional crystal–vacuum interface with a refractive index of $n = 1.8$, the measured resolution is 5.4 μm . This fits well with the diffraction-limited resolution λ/NA for the numerical aperture of $\text{NA} \approx 0.12$ and wavelength $\lambda = 606 \text{ nm}$. However, as the crystal has a length of 1 mm, the limited depth of field at large NA yields an actual resolution limit around 10 μm . Therefore, we use this simple approach only to directly image excitation patterns that are still rather large, while in all other experiments we apply the autocorrelation-based technique for which we then take images at different beam displacements [see Figs. 3(b2) and 3(c2)].

The Stokes beam for STIRAP as well as tripod STIRAP has a width of $w_s = 300 \mu\text{m}$ after passing the telescope consisting of lenses L2 (focal length 75 mm) and L3. It then enters the cryostat counterpropagating to the other beams after being reflected by a D-mirror. This beam line is also used to prepare the initial population distribution in the crystal by optical pumping.

For STIRAP, we use a single-ensemble preparation sequence consisting of burning a spectral pit, repumping, and cleaning [compare Fig. 3(b)]. This results in a spectral antihole with a residual linewidth of about $\Gamma = 2\pi \times 100 \text{ kHz}$ (FWHM). For more information on the preparation sequence, see Ref. [30]. The preparation for tripod STIRAP in Pr:YSO is more tricky [see Fig. 3(c)]. It requires spectral holes around transitions $|g_1\rangle \leftrightarrow |e_1\rangle$ and $|g_3\rangle \leftrightarrow |e_1\rangle$ for the Stokes and control pulses. However, in Pr:YSO it would require a rather complicated class cleaning sequence [33] to prepare only a single ensemble in state $|g_2\rangle$. Thus, for simplicity, we simply burned spectral pits around the relevant transitions and, thus, prepared multiple ensembles. This means that we always simultaneously drive tripod STIRAP in all nine resonant ensembles. Still, by choosing the probe frequency carefully, i.e., by probing the $|g_1\rangle \leftrightarrow |e_3\rangle$ transition, we selectively measure only a single ensemble of interest. We note that this is a specific feature of the inhomogeneously broadened medium Pr:YSO and not of relevance in other media.

V. EXPERIMENTAL RESULTS

We now present and discuss our experimental results on the localization of optical excitations driven by STIRAP, double STIRAP, and tripod STIRAP in Pr:YSO. For the STIRAP and double STIRAP measurements, we applied pulse sequences with Gaussian temporal shapes. The pump and Stokes pulses had durations of $t_{p,s} = 25 \mu\text{s}$ (FWHM) and were truncated within a 125 μs time window. The pulse delay was set to the optimal value (equal to the pulse duration) [20], i.e., $\tau = t_{p,s} = 25 \mu\text{s}$, with the Stokes pulse preceding the pump. For tripod STIRAP, we shortened the pulses to $t_{p,s} = 10 \mu\text{s}$ (FWHM), used a corresponding delay of $\tau = 10 \mu\text{s}$, and truncated them at 50 μs to ensure temporal overlap with the rectangular control pulse of duration $t_c = 100 \mu\text{s}$. The probe pulse had a duration of 10 μs (FWHM) and was truncated at 50 μs . To avoid perturbing the population distribution, the peak probe Rabi frequency was set to $\Omega_{\text{pr}} = 2\pi \times 10 \text{ kHz}$. We typically averaged between 60 and 100 times for a single data point.

A. Validation of autocorrelation measurements

To ensure that our autocorrelation-based measurement technique yields correct localization widths, we validated it for excitation regions with sufficiently large diameters that could also be directly imaged via probe beam absorption. We compared the localization widths extracted from absorption images with those obtained from autocorrelation measurements. In these experiments, we used STIRAP and double STIRAP to localize the excitations and varied the parameter $R = \Omega_p^2/\Omega_s^2$ up to 120, with $\Omega_p = 2\pi \times 3.3 \text{ MHz}$. Figure 4 shows the results obtained with the different measurement and evaluation techniques.

First, we determined the widths of the excitation regions for different values of R from direct absorption imaging (see diamonds in Fig. 4). For increasing R , the width appears to converge toward a minimum value of 10 μm (FWHM), which is simply due to the resolution limit of our imaging system. Therefore, STIRAP and double STIRAP also appear to yield

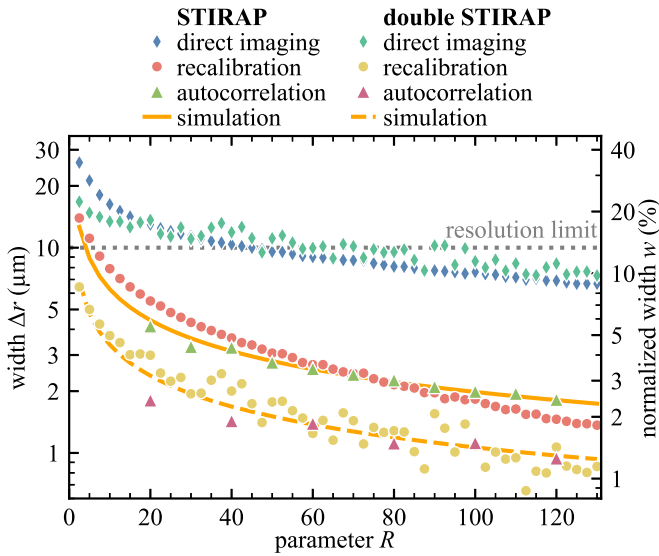


FIG. 4. Localization widths Δr for STIRAP and double STIRAP versus the parameter $R = \Omega_p^2/\Omega_s^2$, determined using different measurement and evaluation techniques. These measurements were taken for beam diameters still well above the wavelength. (i) Experimental data for STIRAP (blue diamonds) and double STIRAP (green diamonds) obtained from direct absorption imaging of the probe beam profile. The gray dotted line indicates the 10 μm two-point resolution limit of our imaging setup. (ii) After recalibration of the direct-imaging data, we obtain the data points shown as red and yellow dots. (iii) Localization widths for STIRAP (green triangles) and double STIRAP (purple triangles) obtained from autocorrelation measurements. (iv) Comparison with numerical simulations for STIRAP (orange solid line) and double STIRAP (orange dashed line). The Stokes beam was set to a Rabi frequency of $\Omega_s = 2\pi \times 300$ kHz with a beam waist of $w_s = 300$ μm . The pump beam had a waist of $w_p = 75$ μm , and we increased the Rabi frequency up to $\Omega_p = 2\pi \times 3.3$ MHz, yielding a maximum value of $R = 120$.

the same performance. A simple calculation (solid and dashed orange lines) based on Eq. (6) confirms that the region of spatially confined excitation should be much smaller than the absorption data indicate at first glance. This limited resolution becomes very apparent when we inspect the images of the atomic populations obtained from absorption measurements after STIRAP [see Fig. 5 (first row)]. The images are strongly blurred, and stronger localization at larger values of R only further flattens the profile. As we have shown in previous work [12], it is nevertheless possible to extract information about the true width of the confinement even from such blurred absorption profiles using a “recalibration” procedure, which works as follows: As R increases, the imaged population peak no longer appears to become narrower; instead, it merely becomes dimmer, i.e., the peak signal and the integral over the peak decrease. The more effectively STIRAP spatially localizes excitation, the fewer quantum systems are addressed in the medium and contribute to the overall signal. Consequently, the total integrated signal scales with the number of systems selected by STIRAP and is independent of the spatial resolution of the imaging setup. If the measured width is limited by the imaging system, this manifests as a reduced signal amplitude. This relationship allows us to infer the ac-

tual width of the localization pattern even when the data are resolution-limited. To do so, we rescale curves fitted to the experimental data such that their peak value is normalized to one, while simultaneously narrowing them to keep the integral constant. For more details, see Ref. [12]. We apply this recalibration method to the data obtained with direct absorption imaging (see the dots in Fig. 4). The widths extracted from the resolution-limited absorption measurements are now much closer to the theoretical prediction. For conventional STIRAP, some deviations are likely due to the fact that the integral is only approximately constant, because there is residual laser power in the dark center of the donut beam. For the recalibrated double STIRAP data, we also observe larger fluctuations due to very small integral values at strong localization (see Fig. 5, third row). In this case, noise becomes much more relevant, which also limits the applicability of the recalibration procedure. Nevertheless, the localization widths obtained from the recalibrated absorption measurements agree sufficiently well with theory to permit comparison with our autocorrelation measurements.

For STIRAP autocorrelation, at each value of R , we recorded multiple images of the resulting excitation profiles for different beam displacements d to reconstruct the autocorrelation function. From this, we inferred the original profile and its width (see Sec. III). The resulting data are shown as triangles in Fig. 4. They agree very well with theory—much better than the widths obtained from direct imaging—because the autocorrelation technique is not resolution-limited. In these initial measurements, the autocorrelation data enable us to determine spatial confinements down to 1 μm , i.e., an order of magnitude below the resolution limit of our imaging system. The green lines in Fig. 5 (second and fourth rows) show cuts through the two-dimensional excitation profiles. In particular, for double STIRAP (see Fig. 5, fourth row), the localization lies far below the resolution limit, such that the distributions appear essentially flat for all values of R . However, using autocorrelation, we recover the actual population distribution (see blue lines in Fig. 5), which matches the numerical simulation (orange lines) almost perfectly. This clearly demonstrates that our autocorrelation technique yields valid results. Already in these test measurements, we achieved localization of optical excitation down to 1.8 μm (FWHM) with STIRAP and 0.94 μm with double STIRAP, corresponding to 2.5% and 1.3% of the pump beam diameter ($w_p = 75$ μm). Hence, STIRAP enables spatial confinement of optical excitation far below the diameter of the driving laser beams. Moreover, the data confirm that double STIRAP improves localization by approximately a factor of 2, as expected.

B. Subwavelength localization by STIRAP

To push the limits of STIRAP localization, we reduce the pump beam waist to $w_p = 13$ μm (see Fig. 6, top right). Due to technical constraints and the fact that STIRAP localizes in the ground state, this is the smallest beam size that can be reasonably used in our setup.

We again chose $\Omega_s = 2\pi \times 300$ kHz for the Stokes Rabi frequency and varied the pump Rabi frequency Ω_p from $2\pi \times 1.3$ to $2\pi \times 4.2$ MHz. This yields values of R ranging

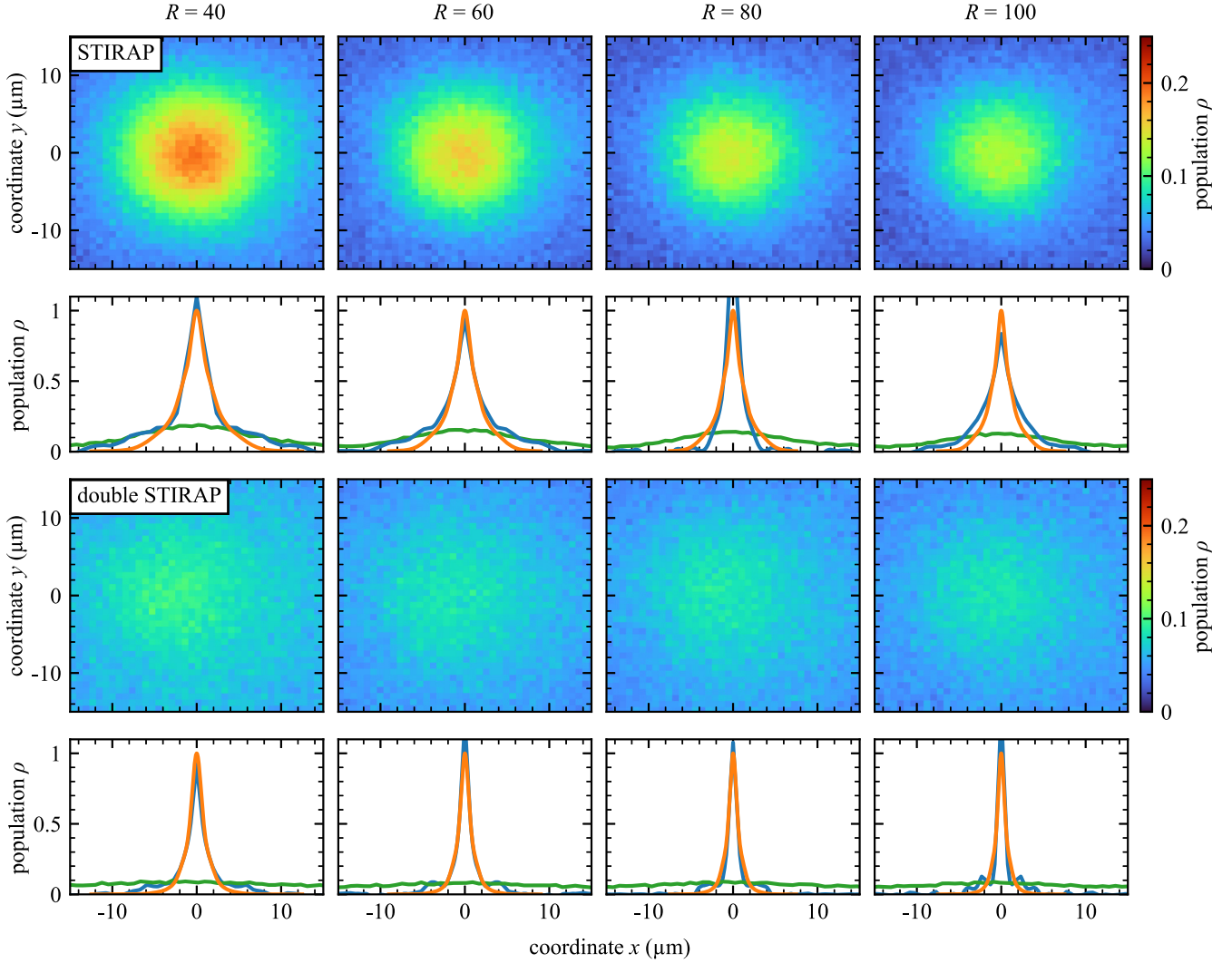


FIG. 5. Spatial variation of the atomic population in state $|1\rangle$ after STIRAP (first row) and double STIRAP (third row) for different values of $R = \Omega_p^2/\Omega_S^2$. The data were obtained from absorption measurements across the probe beam profile. The profiles are rather blurred due to the resolution limit of the imaging system (about $10\ \mu\text{m}$). The second and fourth rows show horizontal cuts through the two-dimensional profiles obtained by direct imaging (green line), compared with the results from autocorrelation measurements (blue line) and a numerical simulation (orange line). The autocorrelation data permit imaging of features far below the resolution limit and agree very well with the simulations.

from 20 to 200 in steps of 10. For each data point, we averaged over 150 images, because at such strong spatial confinement only a small number of quantum systems are addressed and contribute to the signal. For values of $R > 140$, we set the step size of our stepper motor to its minimum of 40 nm and measured up to a maximum displacement of 800 nm. To obtain the autocorrelation function, we summed over a 10×10 pixel ($8\ \mu\text{m} \times 8\ \mu\text{m}$) square in the node of the donut beam. Figure 7(a) shows examples of autocorrelation functions obtained by this procedure (blue triangles). The noise in the data is rather large for small R , but decreases for larger values of R . This is surprising at first glance, but it is likely caused by signal contributions from the ground-state population outside the beam profile due to the finite resolution of our imaging system. This effect is larger when the region outside the beam profile is closer to the node, as is the case for small R . Nevertheless, with an appropriate algorithm (see Appendix A), we can also retrieve autocorrelation functions of sufficient quality

under higher-noise conditions [see the green line in Fig. 7(a)]. Figure 7(b) shows the retrieved population distributions (blue line) after deconvolution of the experimental data via Eq. (15). The results agree remarkably well with numerical simulations (orange line). The amplitudes deviate slightly from unity, but the slopes match very well, in particular at the relevant width defined by $\rho = 0.5$. In Fig. 6, we plot the retrieved localization width (blue triangles) and the theoretical expectation (orange line) as a function of R for a more quantitative comparison. The experimental results agree very well with theory. Most importantly, we find subwavelength localization to a width of $260 \pm 40\ \text{nm}$, i.e., well below our laser wavelength of 606 nm, and roughly two orders of magnitude below the beam diameter as well as the resolution limit of our imaging system. Compared with the beam diameter, this also outperforms the best results obtained so far by related EIT-based localization [22]. The latter work demonstrated a localization width of 60 nm, which is only a factor of about 7 below the

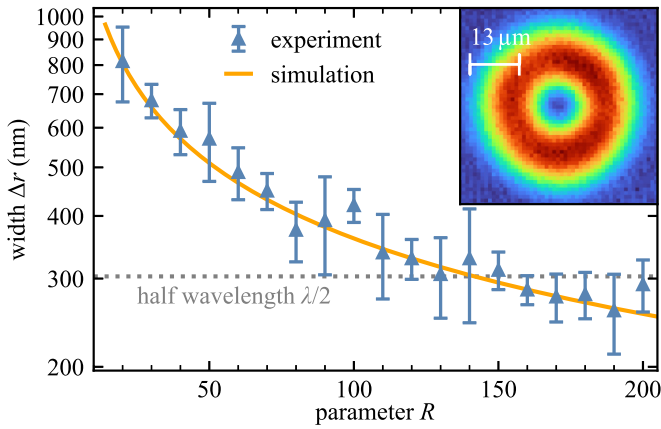


FIG. 6. Localization width Δr for STIRAP as a function of the parameter $R = \Omega_p^2/\Omega_s^2$. The widths are determined from autocorrelation measurements (blue triangles) and compared with a theoretical prediction based on Eq. (6) (orange line). For sufficiently large R , STIRAP reaches a spatial confinement well below half the laser wavelength ($\lambda/2 = 303$ nm; gray dotted line). Note the logarithmic scale on the vertical axis, which improves the visibility of the data points at small localization widths. The inset shows the profile of the donut-shaped pump beam with a waist of $w_p = 13$ μm .

standing-wave periodicity used in the EIT experiment (which corresponds to the beam diameter in our case). Moreover, the standing-wave configuration in Ref. [22] inevitably leads to a periodic array of excitations rather than a single, well-defined excitation spot, as in our approach.

We note, however, that we also only produce spatial confinement in the transversal direction of the laser beam. As for the longitudinal direction, the beam diverges by 0.7 μm within

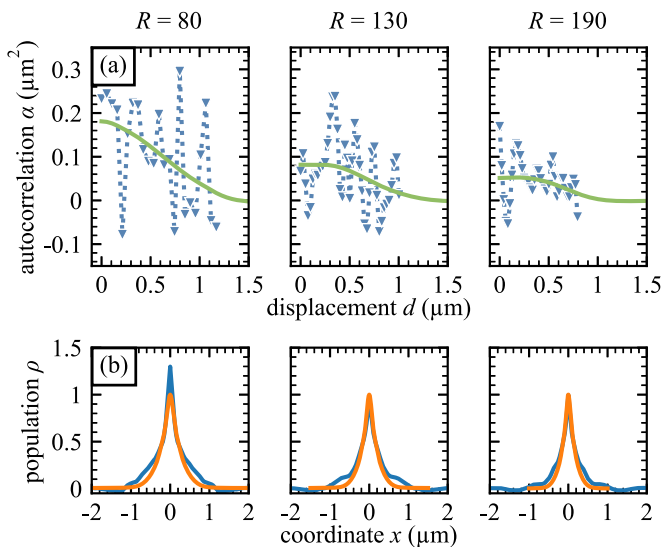


FIG. 7. Autocorrelation measurements to determine subwavelength spatial confinement by STIRAP. (a) Raw data of measured autocorrelation functions α (blue triangles) as well as the continued, interpolated, and filtered data (green line) for different values of $R = \Omega_p^2/\Omega_s^2$. (b) Spatial population profiles reconstructed from the autocorrelation functions (blue line), compared with numerical simulations (orange line).

the 1-mm-long crystal, as the Rayleigh length is 1.5 mm. Therefore, there is almost no variation of beam profiles in the longitudinal direction and, hence, the population distribution in this direction is constant and homogeneous.

We also tried to apply double STIRAP in the setup with a small pump beam diameter to improve the localization even further. However, as double STIRAP improves the localization by a factor of $1/2$, the autocorrelation amplitude shrinks by a factor of $1/4$, while the noise level remains the same. Therefore, we could not confirm the performance of double STIRAP in the subwavelength regime at a sufficient noise level—though of course it is expected to improve the localization of STIRAP as we saw for the larger beam diameter (see Sec. V A).

C. Subwavelength localization by tripod STIRAP

We now proceed to localization by tripod STIRAP. To this end, we modify the experimental setup accordingly [see Fig. 3(a)] to provide pump and Stokes beams with Gaussian profiles and the control beam with a donut-shaped profile.

For a first demonstration, we choose rather large beam waists of $w_p = w_s = 100$ μm and $w_c = 110$ μm , which allows us to directly image the resulting population distributions. The pump and Stokes Rabi frequencies are set to $\Omega_p = \Omega_s = 2\pi \times 200$ kHz, i.e., close to the adiabaticity limit due to power constraints. We vary the control Rabi frequency Ω_c from 0 to $2\pi \times 4$ MHz. Figure 8 shows the resulting localization of optical excitations. The top row shows images of the two-dimensional population distributions in state $|3\rangle$ for different values of Ω_c , and the bottom row shows horizontal cuts through the centers of the distributions (blue line), together with a numerical simulation (orange line). Without a control pulse ($\Omega_c = 0$), we obtain homogeneous, high-fidelity STIRAP transfer across the beam profiles. When we switch on the control pulse and increase Ω_c , the localization shrinks as expected. The measured localizations for larger peak Rabi frequencies ($\Omega_c \geq 2\pi \times 2$ MHz) deviate from the simulation, which is simply due to the 10 μm resolution limit of our imaging system. At higher Rabi frequencies, we obtain stronger spatial confinement, which our imaging system can no longer resolve. We note that, for large Rabi frequencies, a ring-like structure starts to appear around the central peak of localized population transfer. This is caused by off-resonant coupling of the donut-shaped control beam to other transitions in our medium outside the intended tripod STIRAP coupling scheme. When the control Rabi frequency approaches the hyperfine splitting of about 4 MHz in our Pr:YSO crystal, saturation broadening leads to excitation of neighboring transitions. Hence, this is an artifact of our specific medium, which has many closely spaced transitions outside the tripod STIRAP coupling scheme. Nevertheless, the data confirm that tripod STIRAP efficiently localizes coherent population transfer to a target (“excited”) state rather than confining population in the initial state, as in conventional STIRAP.

To push the limits of tripod STIRAP, we reduce the control beam waist to $w_c = 10$ μm (see Fig. 9, top right) and the pump beam waist to $w_p = 4$ μm to ensure full overlap and efficient gating. We keep the Stokes beam waist at $w_s = 300$ μm , as we also require this beam path to optically

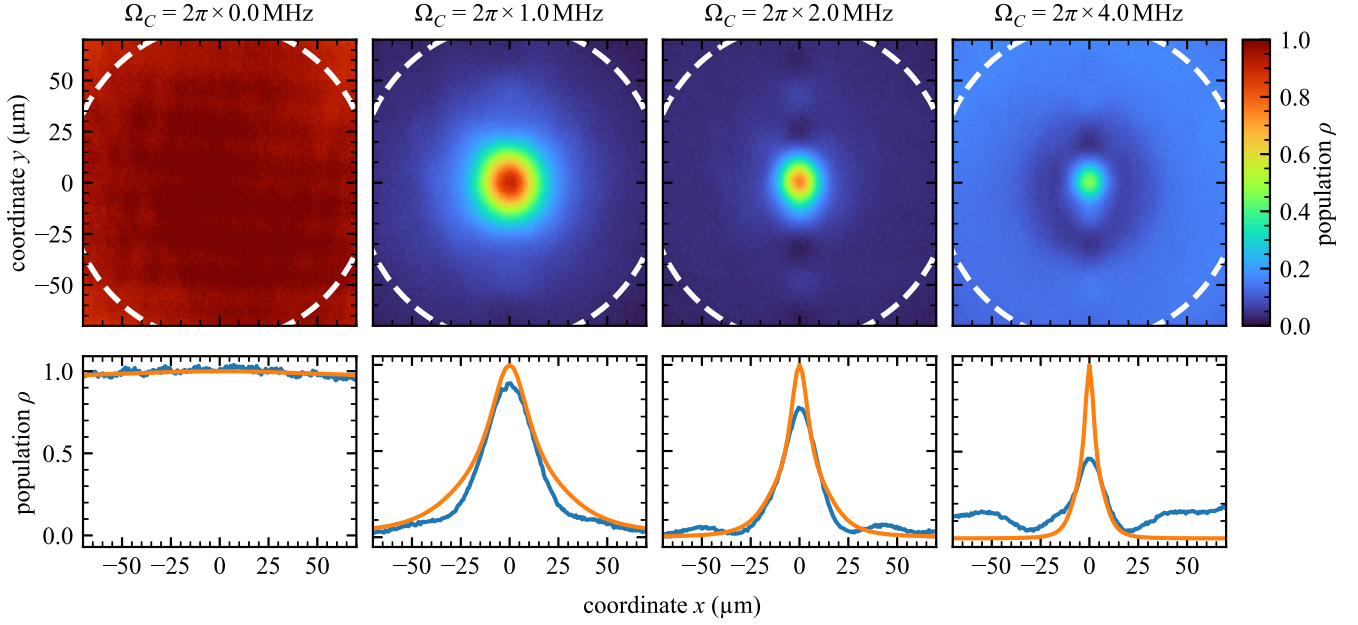


FIG. 8. Spatial variation of the atomic population in state $|3\rangle$ prepared by tripod STIRAP for different peak control Rabi frequencies Ω_C . The data in the top row were obtained by direct imaging of the absorption across the probe beam profile. The white circles indicate the control beam maximum at $w_C/\sqrt{2} = 78 \mu\text{m}$. The lower row shows horizontal cuts (blue line) through the centers of the population profiles, compared with a numerical simulation (orange line). Tripod STIRAP clearly confines optical excitations far below the laser beam diameter.

prepare the crystal. The large Stokes beam diameter compared with the control beam is not problematic for tripod STIRAP because the Stokes beam couples a transition with no initial population; i.e., there is no population transfer in regions where the Stokes beam does not overlap with the control beam. The pump and Stokes Rabi frequencies are set to $\Omega_p = \Omega_s = 2\pi \times 160 \text{ kHz}$. We are limited here by the available laser power and by the need for an additional 10 : 90 (R : T) beam splitter to provide perfectly collinear propagation

of the control and pump beams [see Fig. 3(a)] while maintaining high power in the control beam for efficient gating. Unfortunately, as also predicted by simulation, the limited pump and Stokes Rabi frequencies permit only a transfer efficiency of 92% in the detected ensemble. Nevertheless, this is sufficient for our demonstration. We use the full experimental sequence depicted in Fig. 3(c) to measure localization by tripod STIRAP via the autocorrelation technique. For details on the differences between this measurement and the one for conventional STIRAP, we refer to Appendix B.

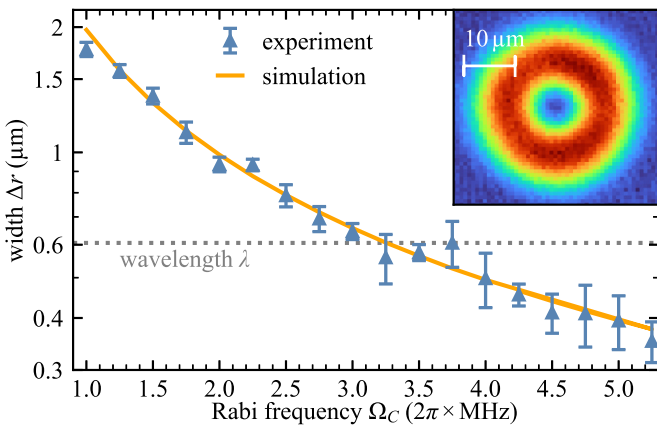


FIG. 9. Localization width Δr for tripod STIRAP as a function of the control Rabi frequency Ω_C . The widths are determined from autocorrelation measurements (blue triangles) and compared with a numerical simulation (orange line). For large Ω_C , tripod STIRAP reaches a spatial confinement well below the laser wavelength ($\lambda = 606 \text{ nm}$; gray dotted line). Note the logarithmic scale on the vertical axis, which improves the visibility of the data points at small localization widths. The inset shows the profile of the donut-shaped control beam with a waist of $w_C = 10 \mu\text{m}$.

Figure 9 shows the experimentally determined localization widths for control Rabi frequencies Ω_C from $2\pi \times 1 \text{ MHz}$ to $2\pi \times 5.25 \text{ MHz}$. We find excellent agreement between the experimental data (blue triangles) and the numerical simulation (orange line). The control-induced spatial gating of STIRAP transfer improves with increasing Ω_C , as expected. For the amplitudes of the recovered population distributions, we obtain the expected value $\rho_{\text{rec}}(\mathbf{r} = 0) = 0.92$ when we choose the low-pass cutoff frequency ν as described in Appendix A. Using tripod STIRAP—even under unfavorable conditions due to limited pump and Stokes power—we spatially confine optical excitation to the target state down to $350 \pm 40 \text{ nm}$, which is well below the laser wavelength of 606 nm (dotted gray line) and about 3.5% of the beam waist. This establishes tripod STIRAP as a favorable alternative to conventional STIRAP and related techniques for localizing optical excitations, with the important advantage of confining population in a target (excited) state.

D. Coherently prepared two-dimensional excitation patterns by tripod STIRAP

Localization of population in a target state (rather than the initial state) enables us to employ tripod STIRAP to prepare

two-dimensional excitation patterns with tightly spaced excitation spots. The idea is to prepare a first spot of excited population with tripod STIRAP and then slightly move the control beam. If we displace the control beam by at least the width of the first localized spot, the pump and Stokes pulses have no effect on the first spot due to efficient gating. Hence, we can generate a second excitation spot at the new position of the control beam node. By repeating the process, we can prepare a two-dimensional grid of tightly confined and closely spaced optical excitations, i.e., population in the target state of the medium. As the ground-state lifetime in Pr:YSO exceeds 100 s at cryogenic temperatures of 4 K, the excitation pattern is long-lived.

For a demonstration experiment, we used our simple imaging setup (without autocorrelation) to directly image the excitation patterns prepared by tripod STIRAP. Due to the limited resolution of the imaging system, we chose relatively large beam waists, i.e., a pump beam waist of $w_p = 18 \mu\text{m}$, a control beam waist of $w_c = 60 \mu\text{m}$, and a Stokes beam waist of $w_s = 300 \mu\text{m}$. We displace the collinear control and pump beams in the horizontal and vertical directions via refraction using two glass plates [see Fig. 3(a)], which are rotated by computer-controlled stepper motors. We set the pump and Stokes Rabi frequencies to $\Omega_p = \Omega_s = 2\pi \times 180 \text{ kHz}$ and the control Rabi frequency to $\Omega_c = 2\pi \times 3 \text{ MHz}$. This yields a localization width of $5 \mu\text{m}$ (FWHM), which is a compromise between the resolution limit of our imaging system and sufficiently tight confinement to permit multiple excitation spots within the control beam diameter. We target excitation spots separated by $15 \mu\text{m}$, which is larger than the width of a single spot but still much smaller than the control beam diameter. We apply the experimental sequence depicted in Fig. 3(c2), but repeat the localization (iv) and motor-displacement (v) steps as often as required to form the excitation grid, and omit step (vi).

Figure 10 shows excitation patterns with $N = 5, 13, 25$, and 41 cells, generated by repeated application of tripod STIRAP while moving the beams from top to bottom and from left to right. The dashed white circles indicate the control beam maximum at $w_c/\sqrt{2} = 42 \mu\text{m}$. Tripod STIRAP clearly permits spatial confinement below the diameter of the driving laser beams and the preparation of tightly confined excitation spots without affecting previously prepared spots. The latter is particularly clear for the image with $N = 5$ spots, where all spots have essentially the same excitation (population) amplitude. We note that the spots are not perfectly aligned with one another. This is due to deviations in stepper-motor operation after we reduced the spatial resolution to increase rotation speed and thereby reduce the measurement time in this proof-of-principle experiment. We also note that the signal amplitudes of the individual spots vary, in particular for larger values of N . This is due to the relatively large diameter of the Stokes beam, which we also require to optically prepare the crystal. The varying peak amplitudes indicate accidental repumping caused by the Gaussian Stokes beam profile spanning the whole grid. If we applied different beam lines for Stokes and optical preparation (which requires more laser power), we could easily get rid of this effect.

We now evaluate the limits of tripod STIRAP to prepare grids of excitation spots in our specific medium and under our

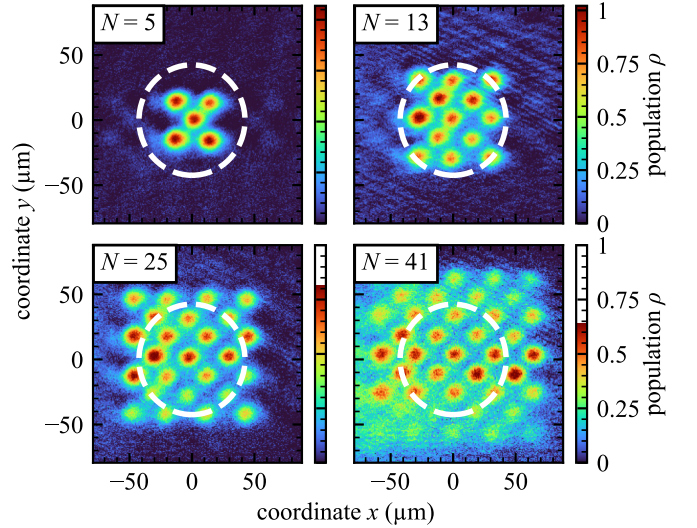


FIG. 10. Excitation patterns prepared by multiple applications of tripod STIRAP with displaced beams. The population in state $|3\rangle$ is measured by absorption across the probe beam profile. We prepared grids with 5, 13, 25, and 41 individual optical excitation spots. The dashed white circles indicate the control beam maximum at $w_c/\sqrt{2} = 42 \mu\text{m}$. Each excitation spot has a width of $5 \mu\text{m}$ (FWHM), and neighboring spots are separated by $15 \mu\text{m}$.

specific conditions of laser parameters. These spots could be applied, e.g., in a quantum memory or a quantum computer. As we demonstrated in the previous section, the technique permits spatial confinements of optical excitation down to roughly 3.5% of the beam diameter (see Fig. 9). Let us suppose a separation between the excitation spots by twice its width. Hence, the spots require a separation of $0.07w_c$. On the other hand, a control beam with a Rabi frequency of $\Omega_c = 2\pi \times 6 \text{ MHz}$ efficiently blocks transfer up to a distance of roughly $1.75w_c$, as we deduced from simulations. Therefore, we can prepare $n = (1.75^2 \pi w_c^2) / (0.07^2 \pi w_c^2) \approx 625$ single excitation spots within the area of the control beam. Assuming that the laser beam diameters could be reduced close to the diffraction limit of about $1 \mu\text{m}$ —which in our demonstration experiment were still large enough to allow direct imaging—the resulting excitation spots could have a width of only 40 nm and be spaced less than 100 nm apart. In media with larger oscillator strengths and fewer perturbing energy levels outside the tripod scheme, these distances could be even smaller. This potential is surely attractive for applications in quantum optics and classical optics.

VI. CONCLUSION

We experimentally demonstrated strong spatial confinement of optical excitations in Pr:YSO well below the laser wavelength using STIRAP as well as double STIRAP and tripod STIRAP which we propose here. For conventional STIRAP, we achieved subwavelength localization of optical excitations down to $260 \pm 40 \text{ nm}$, i.e., only 2% of the pump beam diameter and more than a factor of 2 below the laser wavelength. This is a substantial improvement compared with previous work on STIRAP-driven localization and confirms

the potential of the approach to confine optical excitation far below the diameter of the driving laser.

To measure such small localizations, we developed an autocorrelation-based measurement technique and found excellent agreement with numerical simulations. Using the same method, we also investigated double STIRAP, which, at the expense of coherence, further improves localization by about a factor of 2 to below 1.5% of the pump beam diameter. In principle, this process can be repeated multiple times to further increase the level of spatial confinement. Furthermore, we proposed and implemented tripod STIRAP as a viable coherent technique to tightly localize optical excitations. Due to spatial gating of the adiabatic population-transfer process, tripod STIRAP yields highly localized, high-fidelity population transfer while fully preserving coherence, because the excited state is never populated, unlike in conventional STIRAP. Tripod STIRAP enabled subwavelength localization down to 350 ± 40 nm, i.e., 3.5% of the control beam diameter. This performance falls only slightly short of STIRAP and double STIRAP while significantly outperforming other coherent techniques. Compared with conventional STIRAP, tripod STIRAP localizes atomic population in a target (excited) state rather than in the initial state, which is a major advantage. It enables the creation of two-dimensional patterns of tightly confined atomic populations, as demonstrated by preparing a population grid with up to 41 excitation spots in our crystal without affecting neighboring cells during the excitation process. We emphasize that the present work demonstrates spatially selective, coherence-preserving population transfer. The state-selective readout of an arbitrary state of an individual emitter, and the preservation of arbitrary coherences of spectator emitters during the addressing of a neighboring site, have not yet been characterized and remain an important task for future work.

The implemented techniques provide powerful tools to coherently prepare, address, and manipulate optical excitations on subwavelength scales, with potential applications in quantum technologies and classical optics, e.g., high-resolution storage, addressing, or processing of quantum information, as well as optical microscopy and lithography.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: DETAILED DESCRIPTION OF THE DECONVOLUTION ALGORITHM FOR AUTOCORRELATION FUNCTIONS

Section III already provided a basic description of the deconvolution algorithm, i.e., how to apply the convolution

theorem and Eq. (15) to the measured autocorrelation function $\alpha_{\text{meas}}(d)$ to retrieve the true excitation profile $\rho(\mathbf{r})$. Here, we provide additional details. Figure 11 illustrates the steps of the algorithm.

In Fig. 11(a), we show a measured autocorrelation function (blue triangles). As a first step, to deal with experimental noise, we subtract an offset from $\alpha_{\text{meas}}(d)$ such that the function approaches zero for large displacements d , i.e., when there is no overlap. We then continue the autocorrelation function with zeros to increase the frequency resolution of the Fourier transform, typically up to ten times the maximal displacement. Afterward, we interpolate $\alpha_{\text{meas}}(d)$ with a cubic spline (red dashed line) to resample the data, as displacement via refraction does not directly yield the equidistant samples required for the discrete Fourier transform. Furthermore, this increases the maximal frequency in the spectrum by improving the spatial sampling rate without requiring longer measurements.

Next, we apply a smooth spectral low-pass filter to suppress artifacts. To this end, we use a Butterworth filter with the transfer function

$$G(\omega) = \frac{1}{\sqrt{1 + \left(\frac{\omega}{\nu}\right)^2}}, \quad (\text{A1})$$

with the radial frequency ω and the cutoff frequency ν , which exhibits neither passband nor stopband ripple. Without this filter, the recovered population distribution would exceed the maximum value of unity by an order of magnitude [see gray dash-dotted line in Fig. 12(c)]. We find that even if we convolve the recovered profile with itself again [see gray dash-dotted line in Fig. 12(b)], we reproduce the tail very well but obtain a large deviation at zero displacement. This stems from measuring the one-dimensional autocorrelation $\alpha_{\text{meas}}(d)$ only for positive displacements d and reconstructing the two-dimensional autocorrelation under the assumption of rotational symmetry. The reconstructed function is real and even, so all its Fourier components are zero-phase cosines; any component with nonzero phase must originate from noise and is discarded. Because every cosine peaks at the origin, any surviving contribution—experimental or numerical noise included—interferes constructively at the center, producing the overshoot. The square root in Eq. (15) aggravates this by amplifying noise relative to the signal. Hence, we apply low-pass filtering: the actual localization $\rho(\mathbf{r})$ is bandwidth-limited, and the overshoot is mainly caused by high-frequency noise that interferes constructively in the center. However, care must be taken when choosing the cutoff frequency ν , as the deconvolved localization could otherwise be tuned to an arbitrary width and amplitude. To avoid ambiguity, we deconvolve $\alpha_{\text{meas}}(d)$ using Eq. (15) for various cutoffs ν , yielding reconstructed distributions $\rho_{\text{rec}}(r, \nu)$. We then calculate the corresponding one-dimensional autocorrelations

$$\alpha_{\text{rec}}(d, \nu) = \int_I \rho_{\text{rec}}(r, \nu) \rho_{\text{rec}}(r - d, \nu) dr \quad (\text{A2})$$

for the same displacements d as the measured autocorrelation $\alpha_{\text{meas}}(d)$. Now, to make sure to only filter the additional noise, we check the difference of these reconstructed autocorrelations $\alpha_{\text{rec}}(d, \nu)$ to the original $\alpha_{\text{meas}}(d)$. Ideally, we minimize

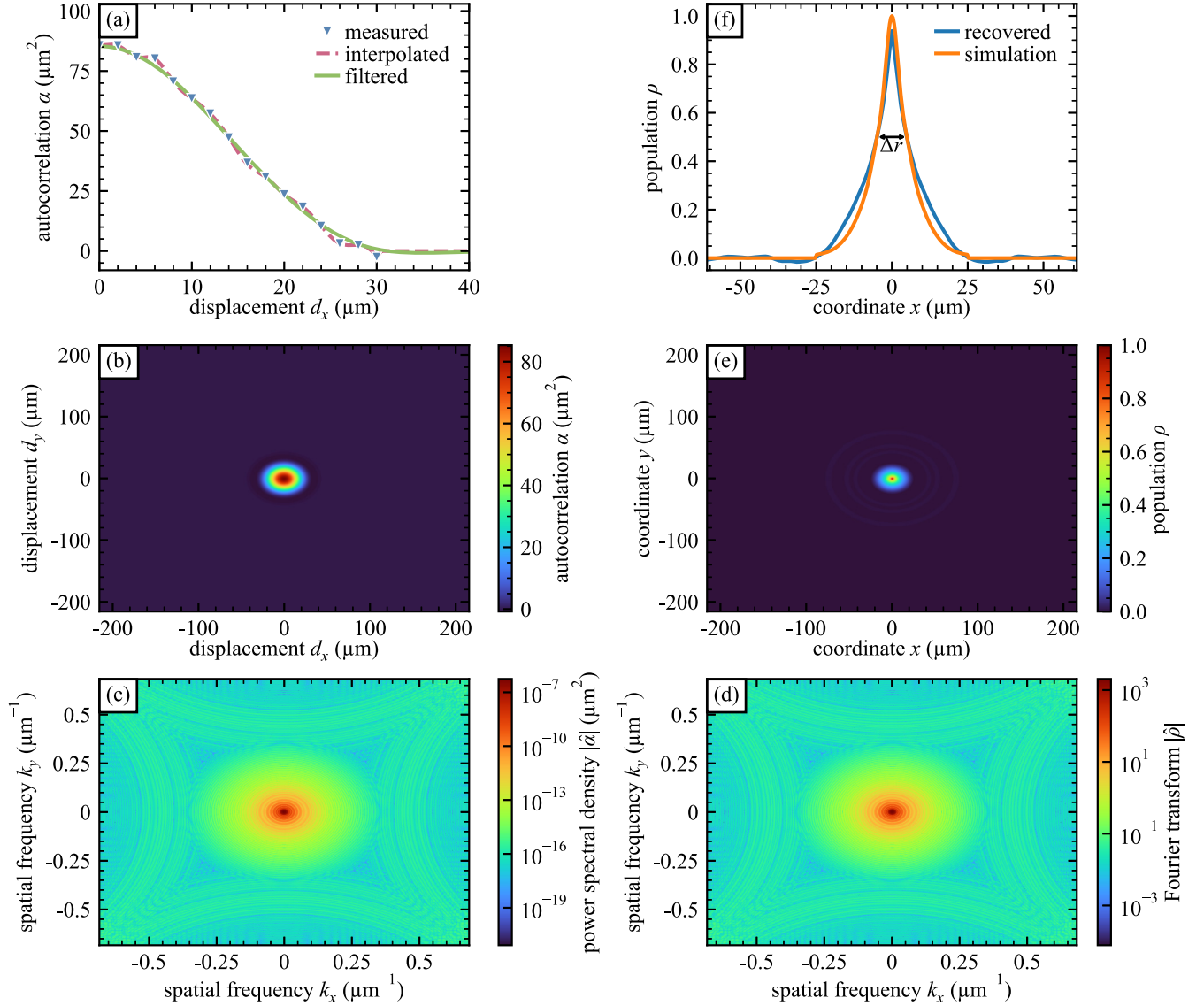


FIG. 11. Steps of the deconvolution algorithm, with piecewise application of Eq. (15) and modifications of the autocorrelation function. (a) Measured one-dimensional autocorrelation function $\alpha_{\text{meas}}(d)$ (blue triangles), the extended version obtained by spline interpolation (red dashed line), and the resulting autocorrelation after applying the low-pass filter (green line). (b) Corresponding two-dimensional autocorrelation function $\alpha(\mathbf{d}) = \alpha(d_x, d_y)$ assuming rotational symmetry, (c) magnitude of the Fourier spectrum of the autocorrelation (power spectral density) $|\hat{\alpha}(k_x, k_y)|$, (d) resulting magnitude of the Fourier transform of the excitation profile $|\hat{\rho}(k_x, k_y)|$ after normalization and taking the square root, (e) two-dimensional population distribution $\rho(x, y)$ after inverse Fourier transformation, and (f) a horizontal cut through the excitation profile $\rho(r)$ (blue line) together with a numerical simulation (orange line). The relevant width Δr at $\rho = 0.5$ is indicated.

the mean square distance (MSD)

$$[\Delta\alpha(v)]^2 = \frac{1}{n} \sum_d [\alpha_{\text{meas}}(d) - \alpha_{\text{rec}}(d, v)]^2 \quad (\text{A3})$$

between the functions with the number of displacements n to find the minimally altered $\alpha_{\text{rec}}(d, v)$. Figure 12(a) shows a plot of the root-mean-square (rms) deviation $\Delta\alpha(v)$ (blue line), i.e., the square root of the MSD $[\Delta\alpha(v)]^2$, as an example. As the choice of the cutoff also affects the maximal amplitude $A(v)$ in the center of the reconstructed population distribution $A(v) = \rho_{\text{rec}}(r = 0, v)$, we can rescale the abscissa from an inconclusive cutoff v to the appropriate amplitude $A(v)$. Thus, we calculate $\Delta\alpha(v)$ for v ranging from a lower boundary v_{min}

(yellow dotted line) with $A(v_{\text{min}}) = 0.6$ to an upper bound v_{max} (red dashed line) with $A(v_{\text{max}}) = 1.8$. We now clearly see a well-defined minimum for an optimal cutoff frequency v_{opt} (green line), which incidentally is also located near the expected maximal amplitude of 1. The corresponding reconstructed autocorrelations $\alpha_{\text{rec}}(d, v)$ are shown in Fig. 12(b), where we clearly see that the unfiltered autocorrelation function significantly overshoots close to the node, while the optimal α_{rec} traces the measured autocorrelation α_{meas} very well. Figure 12(c) also shows the deconvolved population distributions, where the optimal cutoff produces a profile that is close to the expected amplitude.

After we determined the optimal cutoff frequency v for the low-pass filtering of our already recalibrated, continued

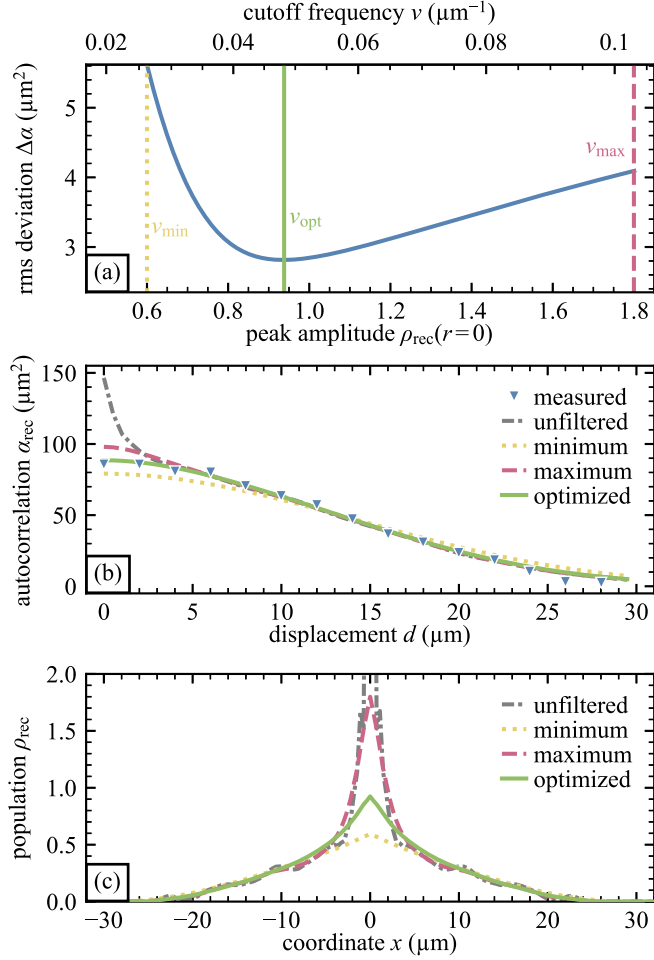


FIG. 12. (a) Root-mean-square (rms) deviation $\Delta\alpha(\nu)$ between the reconstructed and measured autocorrelations, i.e., the square root of the mean square distance (MSD) from Eq. (A3). We choose the minimum at $\rho_{\text{rec}}(\mathbf{r}=0) = 0.94$ as the cutoff for minimal alteration. (b) Recovered autocorrelations $\alpha_{\text{rec}}(d, \nu)$ for three cutoffs ν , as well as the recovered autocorrelation without filtering (gray dashed line), compared with the measured autocorrelation function $\alpha_{\text{meas}}(d)$ (blue triangles) versus displacement d . (c) Excitation profiles versus spatial position for different cutoffs ν . The beam waists were set to $w_p = 160 \mu\text{m}$ for the pump and $w_s = 300 \mu\text{m}$ for the Stokes. The Rabi frequencies are $\Omega_p = 2\pi \times 1.45 \text{ MHz}$ (pump) and $\Omega_s = 2\pi \times 330 \text{ kHz}$ (Stokes).

and interpolated one-dimensional autocorrelation function, we can continue with deconvolution via Eq. (15). By assuming rotational symmetry we reconstruct a two-dimensional autocorrelation function $\alpha(\mathbf{d})$ [see Fig. 11(b)] from the one-dimensional $\alpha_{\text{rec}}(d, \nu_{\text{opt}})$ [green line in Fig. 11(a)], which we just obtained. Then, we perform the discrete Fourier transform [Fig. 11(c)] to obtain the power spectral density $\hat{\alpha}(k_x, k_y)$. Afterward, we normalize and take the square root [Fig. 11(d)], which yields the Fourier transform of the population distribution $\hat{\rho}(k_x, k_y)$. Finally, we take the inverse Fourier transform and obtain the resulting two-dimensional population distribution $\rho(x, y)$ [Fig. 11(e)]. Taking a one-dimensional cut through the center reveals excellent agreement with a numerical simulation [Fig. 11(f)]. The recovered width is then given

as the width Δr of the recovered population distribution at $\rho_{\text{rec}}(r) = 0.5$, as indicated in Fig. 11(f). We use this fixed threshold rather than half of the reconstructed peak amplitude, because the deconvolution can leave the peak slightly above or below unity; the fixed threshold thus makes Δr robust against such amplitude deviations and coincides with the full width at half maximum (FWHM) for the unit-amplitude profile of the idealized theory. To estimate the uncertainty of the recovered width, we characterize each measurement by its signal-to-noise ratio (SNR) $\text{SNR} = \alpha(0)/\sigma_\alpha$, where σ_α is the rms residual between the measured autocorrelation and its filtered version with $\text{SNR} \approx 1.5\text{--}2.5$ for our raw subwavelength data. Propagating σ_α through the averaging, the square root, and the $\rho = 0.5$ crossing yields a relative width uncertainty of about 15%, e.g., $\sigma_{\Delta r} \approx 40 \text{ nm}$ at a recovered width of 260 nm, which is consistent with the 40 nm minimum displacement step of our setup. A Monte Carlo analysis with synthetic Gaussian profiles of known width confirms that the extraction is unbiased near our operating point at $\text{SNR} \approx 2$, with a residual scatter scaling as $1/(\text{SNR} \sqrt{N})$. The error bars shown on the experimental data in Figs. 6 and 9 represent this uncertainty. The algorithm works the same for any of our measured autocorrelation functions, no matter if they are obtained with STIRAP, double STIRAP, or tripod STIRAP.

APPENDIX B: AUTOCORRELATION MEASUREMENTS FOR TRIPOD STIRAP

As mentioned in Sec. III, the autocorrelation measurements require some modification for tripod STIRAP. This is because we localize in the excited state for tripod STIRAP rather than in the initial state, as is the case for conventional STIRAP. This means that there is no way to clean “residual” population before the second transfer process in the autocorrelation measurement, and we inevitably obtain some transfer from state $|3\rangle$ back to $|1\rangle$ via bright STIRAP [20], which needs to be accounted for. Therefore, the transfer process is bidirectional in tripod STIRAP, whereas for conventional STIRAP the cleaning step ensures that it is unidirectional. This means that the measured population distribution $\rho_{\text{meas}}(\mathbf{r}, \mathbf{d})$ after the second tripod STIRAP transfer with displacement $\mathbf{d}$ is

$$\rho_{\text{meas}}(\mathbf{r}, \mathbf{d}) = \rho(\mathbf{r}) - \beta\rho(\mathbf{r})\rho(\mathbf{r} - \mathbf{d}) + [1 - \rho(\mathbf{r})]\rho(\mathbf{r} - \mathbf{d}), \quad (\text{B1})$$

which does not directly yield the autocorrelation function after integration as for a unidirectional transfer. Here, $\rho(\mathbf{r})$ describes the population distribution after the first transfer and $\rho(\mathbf{r} - \mathbf{d})$ the distribution after the second displaced transfer. The coefficient β takes into account that bright STIRAP might be less efficient than dark state STIRAP [where the efficiency is included in the resulting distribution $\rho(\mathbf{r})$]. The first term in Eq. (B1) describes the population distribution in state $|3\rangle$ generated by the first tripod STIRAP sequence. From this population, we subtract the population that the second sequence transfers from state $|3\rangle$ back to $|1\rangle$ by bright STIRAP. Finally, the third term represents the population that was not transferred by the first sequence but is then transferred by the second after displacement. We show examples of the resulting population distributions $\rho_{\text{meas}}(\mathbf{r}, \mathbf{d})$ in Fig. 13(a).

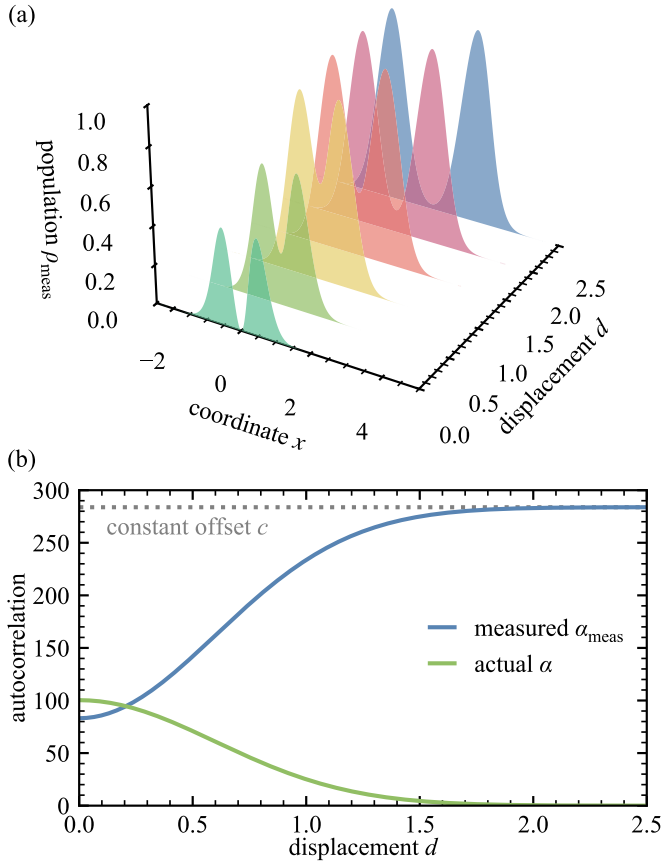


FIG. 13. (a) Measured spatial population distribution ρ_{meas} [see Eq. (B1)] after an initial bidirectional population transfer $\rho(r)$ and a second, displaced transfer $\rho(r - d)$ at various displacements d . Both transfers are assumed to yield a Gaussian-shaped population distribution with a full width at half maximum (FWHM) of 1. (b) Measured autocorrelation function α_{meas} (blue line), corresponding to the integral over the population distributions in panel (a). The measured autocorrelation increases with displacement and converges to a constant offset c , equal to the area under two fully separated population peaks [see the blue profile in panel (a)]. Using Eq. (B3), we reconstruct the actual autocorrelation function α (green line). It decreases with displacement, as expected, and yields the original profile $\rho(r)$ after deconvolution with Eq. (15).

One can clearly see that the resulting populations have peculiar shapes, as the transfers in both directions influence each other. Rearranging Eq. (B1) and integrating over the relevant part of the image I yields the measured autocorrelation function [see blue line in Fig. 13(b)]

$$\alpha_{\text{meas}}(\mathbf{d}) = \iint_I \rho(\mathbf{r}) + \rho(\mathbf{r} - \mathbf{d}) d\mathbf{r} - (1 + \beta)\alpha(\mathbf{d}), \quad (\text{B2})$$

where we identify the actual autocorrelation function $\alpha(\mathbf{d}) = \iint_I \rho(\mathbf{r})\rho(\mathbf{r} - \mathbf{d}) d\mathbf{r}$. We see that the measured and the actual autocorrelation function only differ by a sign and an offset $c(\mathbf{d}) = \iint_I \rho(\mathbf{r}) + \rho(\mathbf{r} - \mathbf{d}) d\mathbf{r}$ (gray dotted line). Note that the integral $\iint_I \rho(\mathbf{r} - \mathbf{d}) d\mathbf{r}$ is independent of $\mathbf{d}$, provided that we integrate over the whole displacement range such that the offset $c(\mathbf{d})$ is indeed a constant c . Assuming we displaced the beam sufficiently far by some maximal displacement $\mathbf{d}_{\text{max}}$, the autocorrelation function should be zero, i.e., $\alpha(\mathbf{d}_{\text{max}}) = 0$, because there is no overlap between the localized peaks anymore [see the blue profile in Fig. 13(a)]. This only leaves the contribution from the offset $\alpha_{\text{meas}}(\mathbf{d}_{\text{max}}) = c$ as the integral over two fully separated peaks, so that, by rearranging Eq. (B2), we find that

$$\alpha(\mathbf{d}) = \frac{\alpha_{\text{meas}}(\mathbf{d}_{\text{max}}) - \alpha_{\text{meas}}(\mathbf{d})}{1 + \beta} \quad (\text{B3})$$

yields the actual autocorrelation function $\alpha(\mathbf{d})$ [see green line in Fig. 13(b)]. The resulting $\alpha(\mathbf{d})$ then decreases with increasing displacement $\mathbf{d}$, as expected. We typically assume bright STIRAP to be as efficient as dark state STIRAP, i.e., we set β to unity. This is justified, as we ensure adiabaticity with sufficiently large pulse areas and the duration of the transfer process stays well below T_1 and T_2 of the excited state $|e\rangle$. Finally, we plug the recovered $\alpha(\mathbf{d})$ into Eq. (15) for deconvolution after the modifications described in Appendix A, as we do for conventional STIRAP.

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