

Spatially localized optical excitations in a doped solid using robust broadband composite pulsesNiels Joseph ^{1,*}, Nikolay V. Vitanov ², and Thomas Halfmann ^{1,†}¹*Institute for Applied Physics, Technical University of Darmstadt, Hochschulstr. 6, 64289 Darmstadt, Germany*²*Center for Quantum Technologies, Department of Physics, Saint Kliment Ohridski University of Sofia, 5 James Bourchier Boulevard, 1164 Sofia, Bulgaria*

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We experimentally demonstrate spatial confinement of optically driven atomic excitation by broadband composite pulses in a rare-earth ion-doped $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal. The experimental data confirm that such sequences enable spatially localized excitation well below the diameter of the applied laser beam, with a substantial improvement compared to previous developments and applications of other classes of composite pulses. In particular, we derived and applied broadband composite sequences, which also work efficiently in inhomogeneously broadened media, while also maintaining robustness with regard to fluctuations of the driving laser intensity. We perform extensive measurements to assess and compare the performance of several classes of broadband composite sequences. The findings are confirmed by numerical simulations, which agree very well with the experimental data. As the concepts are also applicable to spatially confined excitations below the diffraction limit of the laser beam, the results serve as an important step towards high-resolution imaging, preparation and addressing in quantum technologies and classical applied optics.

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Diffraction imposes a fundamental limit for the spatial resolution of light–matter interactions, as it restricts the spatial confinement of optical excitation to approximately half the wavelength of the driving laser beam. Overcoming this limit is crucial for a wide range of applications in classical optics, e.g., high-resolution microscopy [1–6] or nano-scale lithography [7–11], but also in quantum technologies. In the latter field, novel approaches to overcome the diffraction limit are essential to enable high-resolution, coherent addressing, and manipulation of individual qubits in dense ion traps or optical lattices [12–17], for spatially multiplexed quantum memories [18–20], or to enhance light–matter interactions in solid-state media [21]. In classical optics, powerful methods such as stimulated emission depletion (STED) microscopy [1] have been employed to achieve sub-diffraction resolution. However, these approaches usually require multilevel schemes, multiple laser beams, and also rely on incoherent processes. The latter renders them not applicable in quantum information storage or processing, which require preservation of quantum coherence.

One approach to address this challenge is based on composite pulse (CP) sequences. The basic idea of CPs is to drive a quantum system on specific pathways through Hilbert space, which offer robustness versus variations in the interaction

parameters. The relative phases of the single pulses in the composite sequence serve as control parameters to follow such robust pathways. The larger the number of pulses N in the CP sequence, the larger the number of control parameters and the better the performance. Originally developed in NMR spectroscopy [22,23], CPs also attracted attention for applications in quantum technology due to their ability to compensate for fluctuations in the experimental parameters of coherent interactions between radiation fields and a quantum system—which enables precise manipulation of qubits also when the experimental conditions vary. This makes them also applicable to control interaction processes in the spatial domain, e.g., to spatially extend or confine optical excitation probabilities across a laser beam profile. Of particular interest here were narrowband CPs (NCPs). They are called “narrowband” because of their high sensitivity with regard to variations of experimental parameters and, hence, by design exhibit a steep intensity dependence of the excitation probability. Therefore, they allow spatial confinement of excitations far below the laser beam diameter, potentially even below the diffraction limit [16,24,25]. However, NCP sequences converge rather slowly with the number of pulses N and, hence, very long sequences are required to obtain satisfactory results—which is a problem, if the total pulse duration approaches the finite coherence time of a medium. We also note that the performance of NCPs to localize atomic excitations typically falls well behind adiabatic techniques such as STIRAP [11,19], though (as a disadvantage) the latter requires a multilevel scheme and two laser beams.

While NCPs are meant to work efficiently in a small parameter range, broadband composite pulses (BCPs) are designed to maintain large excitation efficiency over a wide range of interaction parameters, e.g., detunings from atomic resonances and fluctuations in the driving laser intensity [24].

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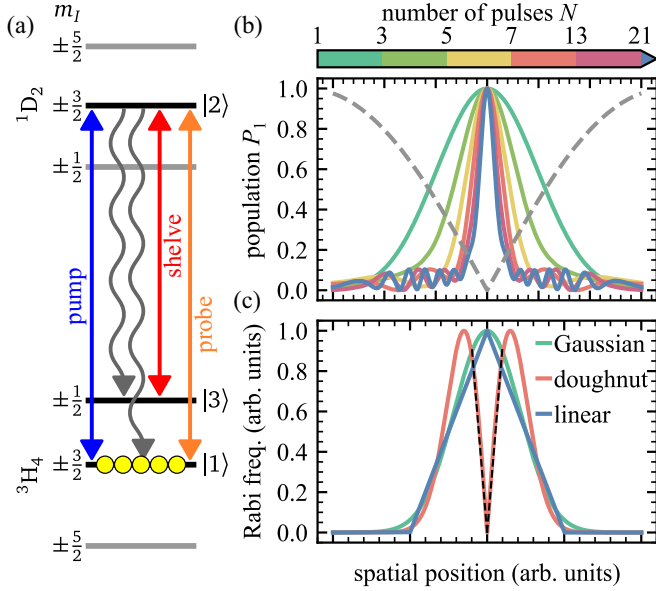


FIG. 1. (a) Coupling scheme in Pr:YSO. Straight lines depict transitions coupled by a laser, wavy lines show spontaneous decay. Yellow circles show the initial population distribution. (b) Numerical simulation of the excitation probability. The plot shows the variation of population P_1 in the initial state $|1\rangle$ after excitation by a BCP sequence (developed in this work) vs spatial position across a doughnut-shaped pump laser beam profile for different numbers of pulses N in the sequence (color code). For comparison, the gray dashed line shows the laser beam profile, given by the spatially varying Rabi frequency. The peak pulse area at the maxima of the beam is set to $A = \pi$. (c) Comparison of different beam profiles, given as spatially varying Rabi frequencies. The black lines indicate a linear approximation of the doughnut profile close to the node.

Yet, their potential to spatially confine atomic excitations remained unexplored. In particular, there are no such investigations in solid-state systems like, e.g., rare-earth ion-doped crystals, which are promising candidates to implement an optical quantum memory with long coherence time, scalability, and relative ease-of-use [26].

In this work, we experimentally demonstrate spatial confinement of atomic excitations by specifically designed CP sequences, driven by a single fixed-frequency amplitude- and phase-modulated doughnut-shaped laser beam in a simple two-level system in a $\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal (hereafter referred to as Pr:YSO). In particular, we developed and implemented several classes of BCPs that substantially improve spatial resolution compared to previous attempts with NCPs [25], and are applicable even in media with inhomogeneous broadening. Our results confirm that BCPs permit strongly localized excitations well below the diameter of the driving laser beam, thereby opening up broad opportunities for applications.

II. BASIC THEORY

In the following, we will briefly summarize the concept of spatial confinements of atomic excitation by broadband composite sequences. We consider a two-level system [see Fig. 1(a)], with the population initially being in the ground state $|1\rangle$. Optical pulses from a pump laser beam resonantly

couple this state to the excited state $|2\rangle$. We describe the laser pulses by a time-dependent pump Rabi frequency $\Omega(t)$. The probability of excitation to state $|2\rangle$ is given by $P_2 = \sin(A/2)^2$, where the pulse area is defined as $A = \int \Omega(t) dt$. For pulsed excitation with peak pump Rabi frequency Ω_p and duration τ , the pulse area scales as $A \propto \Omega_p \cdot \tau$. When the pulse area is equal to π , we get complete population transfer to the excited state, i.e., $P_2 = 1$. This is well-known population inversion by a π pulse. However, we must also consider the spatial dependence of the Rabi frequency across the laser beam profile, which leads to a variation of the excitation probability across the beam profile.

As a simple example, let us consider a Gaussian beam profile, expressed in terms of the spatially varying pulse area $A(r) = \pi \exp(-r^2/w^2)$, with the beam waist w . In the center we have a π pulse, while in the wings the pulse area decreases with the radial coordinate r . Therefore, the optical excitation profile on the $|1\rangle \leftrightarrow |2\rangle$ transition will mirror the Gaussian-shaped beam profile. Indeed, the width of the excitation profile as expressed in terms of the full-width-at-half-maximum (FWHM) is equal to the width of the incident laser beam also given in terms of FWHM. In other words, the width of the excitation profile is typically limited by the laser beam diameter and cannot be reduced further (if we assume excitation by a single laser beam in a two-level system).

To overcome this limit for coherent excitation, in the CP approach we replace the single π pulse by a train of N π -pulses with appropriately chosen relative phases $(\phi_1, \phi_2, \dots, \phi_N)$, which serve as control parameters to tailor the excitation profile. In the case of NCP sequences (developed and investigated in previous work [25]), the goal was to reduce the diameter of the region with large population transfer around $A = \pi$ in the center of a laser beam profile. This can be imagined as destructive interference of the transfer amplitudes in the wings of the profile. Thus, with a NCP sequence we aim to increase the sensitivity of the transfer efficiency $P_2(A)$ vs the pulse area A . Hence, the transfer is narrowband with regard to the pulse area. Therefore, population transfer concentrates around the center of the beam profile [25].

The best spatial confinement demonstrated with NCPs so far reached about 25% of the incident laser beam width and equally the localization achieved with a simple π pulse, i.e., well below the laser beam diameter.

Further investigation in the latter work revealed that the limiting factor for the spatial confinement by NCP sequences is the Gaussian beam profile of the driving laser. The dependence of the Rabi frequency $\Omega(r)$ upon the radial coordinate r is rather flat close to the center of a Gaussian beam profile. Thus, the variation of the excitation probability around the center is flattened as well—which limits the achievable spatial confinement by the NCP sequence. It would be better to have a steeper slope in the beam profile. As an example, see a beam profile with linear slope in Fig. 1(c), which is obviously much narrower compared to a Gaussian. However, such a spatial profile is not possible as solutions to the wave equation have to be quadratic in r .

It turns out that the slope close to the central node of a doughnut-shaped TEM 01^* mode varies approximately linearly with the radial coordinate [see black line in Fig. 1(c)].

Simulations confirm that a linear slope would lead to a quadratic improvement of the spatial confinement when compared to a Gaussian. Although this is an interesting feature, the linearity is close to an intensity minimum rather than a maximum. Therefore, one cannot apply a NCP sequence with a doughnut-shaped beam, as there is no excitation in the node at zero intensity.

Suppose now, we would try a single pulse from a doughnut-shaped beam with the intensity in the maxima corresponding to a pulse area of $A = \pi$. In this case, we get full population transfer to the excited state at the maxima, less population transfer in the outer wings of the profile and the linear slopes next to the center node, and no excitation right in the center node of the beam. Hence, the excitation pattern (i.e., population excited to state $|2\rangle$) across the beam profile would roughly mirror the doughnut beam profile. In turn, if we consider the population left in the ground state $|1\rangle$, we get roughly an inverted image of the doughnut profile with a broad central peak [see turquoise line in Fig. 1(b)].

When we invert this population distribution afterwards by, e.g., a simple π pulse from a sufficiently wide Gaussian-shaped laser beam, we get strongly localized excitation in the center. This could then be mapped from an optical excited state that decays onto a metastable state $|3\rangle$ by an additional shelving beam [see Fig. 1(a)]. Then, one just has to improve the efficiency of the initial doughnut-shaped transfer to reduce the width of the central region where population remains in the ground state $|1\rangle$.

Hence, we require a CP sequence that also enables efficient excitation in the linear slopes next to the center node, where the pulse area is $0 < A < \pi$. Thus, we want a CP sequence which is robust with regard to deviations of the pulse area from π , i.e., a broadband sequence (BCP) rather than a NCP sequence. The more pulses there are in a BCP sequence, the more robust it is, the larger the transfer efficiency next to the node, and consequently the stronger the spatial confinement of population left in ground state $|1\rangle$ [see color-coded lines in Fig. 1(b)].

The application of BCPs has some additional advantages. Firstly, the localization width with BCPs is substantially smaller than for NCPs with a Gaussian profile, as shown in previous theory work [24]. Secondly, there is no need to set the pulse area in the maxima of the beam exactly to π , as BCPs are robust to variations in the pulse area. This is much unlike NCPs, where a slight misalignment from the peak value $A = \pi$ in the center of the beam results in a massive drop of fidelity. As we will show below, this feature of BCPs can even be exploited to further improve the localization capabilities.

However, as an important issue for applications, the BCP sequences developed in previous theory work [24] are sensitive to variations of the detuning from exact resonance in the two-level scheme and, therefore, are also affected by inhomogeneous broadening in a medium. Figure 2 illustrates this behavior. The figure shows different landscapes of excitation, i.e., the variation of the population P_1 after BCP sequences vs pulse area A and detuning Δ . For plot (a) we applied broadband CP sequences consisting of π pulses proposed in [24], which we from now on term BCP type I. Plot (b) shows the behavior of so-called “ultra-broadband”

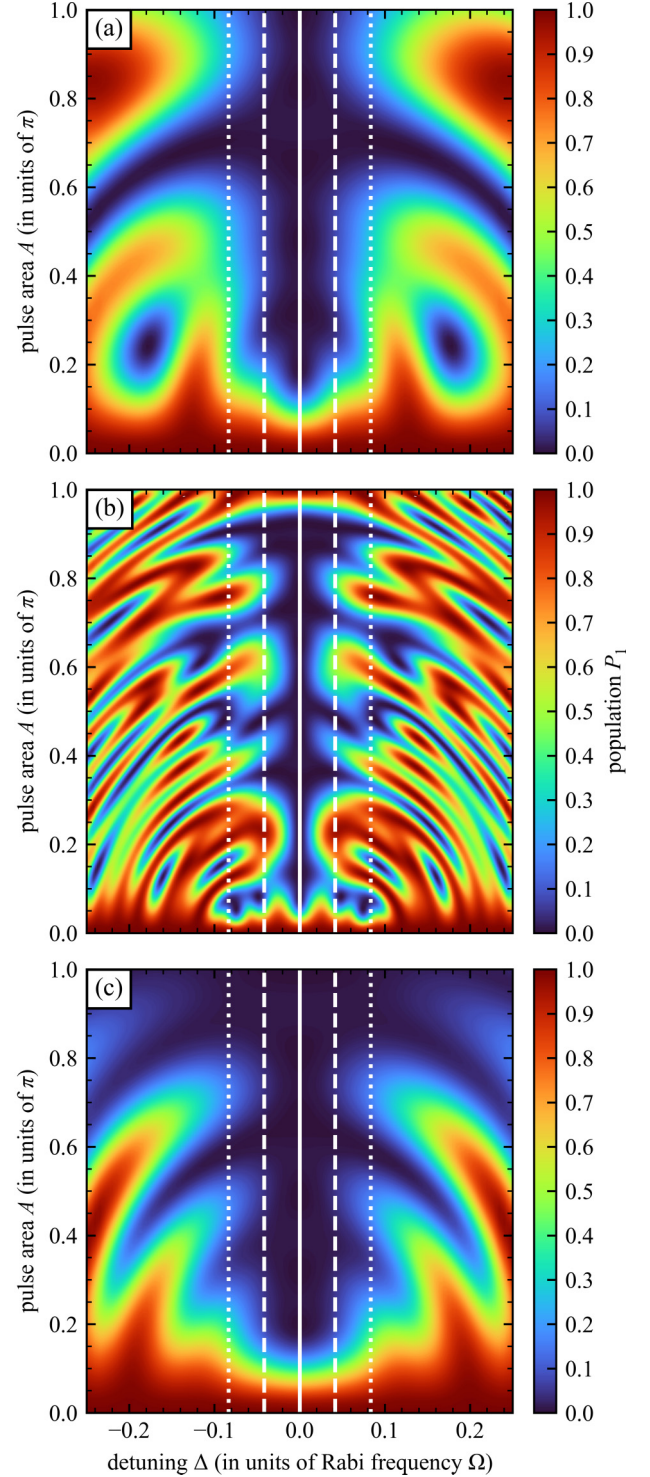


FIG. 2. Numerical simulation of the excitation probability P_1 vs detuning Δ and pulse area A for different BCP sequences. (a) BCP type I with $N = 11$ pulses, (b) BCP type II with $N = 7$ pulses, and (c) IHO BCP with $N = 11$ pulses. The peak Rabi frequency is $\Omega_p = 2\pi \times 1.2$ MHz. The white, dashed lines indicate a spectral linewidth of $\Gamma = 2\pi \times 100$ kHz (FWHM) for inhomogeneous broadening in our system. The dotted lines indicate a detuning range of twice that width.

CP sequences (also developed in [24] as an alternative to BCP type I), where in addition to the phases, the pulse areas also serve as a control parameter. We will refer to them as BCP type II. Because of the addition of more parameters these sequences are even more robust (ultra-broadband, as termed in [24]) to variations in the pulse area. In plot (c), we apply our newly developed sequences that are robust also with regard to inhomogeneous broadening. Therefore, we will call these sequences inhomogeneous-optimized or IHO BCPs, for short.

Let us first consider the case of exact resonance $\Delta = 0$, i.e., no variations of the detuning, hence no inhomogeneous broadening (see solid white line in vertical direction in the plots). In this case, all BCP sequences efficiently drive population to the excited state [2], even if the pulse area deviates from π . Only in a small region $A \approx 0$ (i.e., close to the node of a doughnut-shaped beam profile), we have $P_1 \approx 1$, where population remains in the ground state [1]. Cuts along the axis $\Delta = 0$ yield the population profiles already shown and discussed in Fig. 1(b). Let us now consider inhomogeneous broadening, i.e., detunings from resonance. The dashed white lines in the plots indicate a spectral linewidth with an assumed inhomogeneous broadening of $\Gamma = 2\pi \times 100$ kHz in the medium (which is typical for our experiments in Pr:YSO). The dotted white lines indicate a detuning range of twice that value. When we follow the lines in vertical direction, we see that the population P_1 for excitation by BCP type I or II [see Figs. 2(a) and 2(b)] shows strong variations or even fast oscillations. The larger the detuning, the more pronounced the oscillations.

In particular, we see that we also have population left in the ground state for large pulse areas. This is because of the additional accumulation of phase $\Phi_{\text{add}} = \Delta \cdot \tau$ during the CP-driven excitation process with detuning Δ and pulse duration τ . To calculate the achievable localization in an inhomogeneously broadened medium we have to average over the broadened spectral line, i.e., a range of detunings and, hence, additional phases. This yields sidebands in the excitation profile, which manifest as ring-like structures around the central peak of spatially confined population—which is obviously a perturbing feature to localize excitation. In particular, BCP type II sequences produce many rings because of their complex path around the Bloch sphere. This is owing to the fact that with BCP type II sequences we apply much higher pulse areas per pulse than $A = \pi$, i.e., we traverse more distance on the Bloch sphere which increases the effects of phase misalignment. But even BCP type I sequences, which apply only π pulses, still have significant excitation close to the inhomogeneous line. We note that for Fig. 2(b), we considered a shorter BCP type II sequence with seven pulses, where the robustness with regard to the pulse area is significantly increased. However, this comes at the cost of less robustness with regard to the detuning. Therefore, we see faster oscillations of the population distribution for off-resonant excitation. Thus, these BCP sequences permit quite strong spatial confinement in the center node of a doughnut beam, but in an inhomogeneously broadened medium also generate strong off-resonant background with lots of rings—which renders them not suitable for inhomogeneously broadened media. These limitations of BCP type I

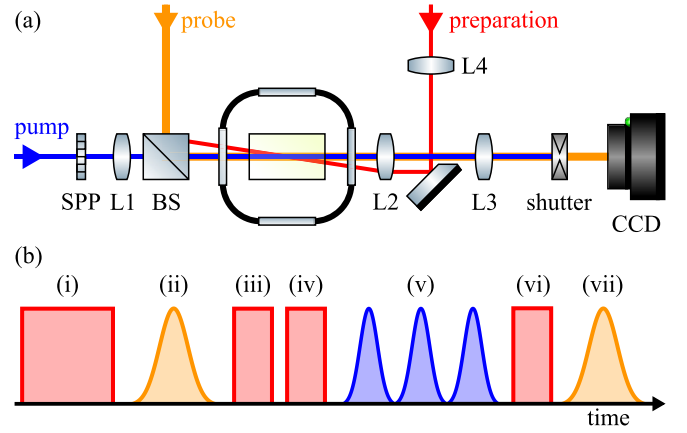


FIG. 3. (a) Experimental setup with pump (blue), preparation (red), and probe (orange) beamlines, Pr:YSO crystal (green), beam splitter (BS), lenses (L), spiral phase plate (SPP), shutter, and CCD camera. (b) Temporal sequence of optical pulses with (i) preparation spectral pit, (ii) probe reference measurement, (iii) repump to generate antihole, (iv) cleaning, (v) CP sequence, (vi) shelving, and (vii) probe measurement. Colors correspond to the beamlines in (a).

and II sequences were not investigated by theoretical work so far [24].

To deal with inhomogeneous broadening, we developed the aforementioned new IHO BCP sequences. To derive these, we added a constraint to the optimization algorithm, which favors sequences suppressing sideband excitation in the spectral region of inhomogeneous broadening. When we compare the landscapes of excitation in Figs. 2(a)–2(c) for BCPs type I and II with our new IHO BCPs, we clearly see the superior performance of the latter, i.e., suppression of sidebands in the full range of detunings corresponding to the inhomogeneous broadening. We note that the theoretical work in [24] derived only CPs with up to $N = 11$ pulses. In our work, we extended the derivations for IHO BCPs up to $N = 25$ pulses. For details on the theoretical treatment and the obtained phases see Appendix.

III. EXPERIMENTAL SETUP

We conduct the experiments in a Pr:YSO crystal, with the $^3\text{H}_4 \leftrightarrow ^1\text{D}_2$ optical transition at a center wavelength of 605.98 nm. Specifically, we use the hyperfine states $|1\rangle = ^3\text{H}_4, |m_I = \pm 3/2\rangle$, and $|2\rangle = ^1\text{D}_2 |m_I = \pm 3/2\rangle$ as a two-level quantum system, manipulated via BCP sequences on a pump beam [see Fig. 1(a)]. The coherence time of the optical transition $T_2^e = 111 \mu\text{s}$ imposes an upper limit on the total duration of the BCP sequences and the maximum number of pulses N they contain.

The experimental setup, shown in Fig. 3(a), is centered around a Pr:YSO crystal (1-mm long, doped at 0.05%), which is cooled to temperatures below 4 K in a continuous-flow cryostat (ST-100, Janis Research Co.). We derive the laser beams for the experiment from an amplified diode laser system with a frequency-doubling unit (DLC TA-SHG pro 24509, Toptica Photonics). This system delivers 800 mW of optical power to the experiment and achieves a frequency stability of about 20 kHz (FWHM).

We use acousto-optical modulators to generate preparation, pump, and probe laser pulses, enabling precise manipulation of their intensity, frequency, and phase. We control the temporal shapes of preparation and probe pulses with waveform generators (33220A, Agilent) in conjunction with direct digital synthesis drivers, while BCP sequences on the pump beam are controlled by an arbitrary waveform generator (AWG5014, Tektronix). This configuration provides a phase accuracy better than $\pi/200$, well within the required $\pi/100$ precision for efficient BCP sequences with several tens of pulses, as confirmed by our numerical simulations. To ensure a proper Gaussian profile in space, the pump beam passes through a spatial filter [not shown in Fig. 3(a)] before passing a spiral phase plate with charge 1 (V-593-20-1, Vortex Photonics), which imprints an azimuthally increasing phase onto the beam. This produces a Laguerre-Gaussian TEM₀₁* doughnut-shaped Rabi frequency distribution $\Omega(r) = \Omega_p \frac{r}{w_p} e^{-r^2/w_p^2}$ with radial coordinate r , maximum Rabi frequency Ω_p , and waist w_p . Afterwards, lens L1 (focal length 200 mm) mildly focuses the beam into the crystal down to a beam waist of $w_p = 160 \mu\text{m}$.

After the BCP sequence, the excited population in state $|2\rangle$ would decay with a lifetime of $T_1^e = 164 \mu\text{s}$, primarily into states $|1\rangle$ and $|3\rangle = {}^3\text{H}_4 |m_I = \pm 1/2\rangle$. However, this would destroy the localized population pattern that we intended to be left in state $|1\rangle$. Therefore, we apply an optical “shelving” π pulse on the preparation beam line to remove the perturbing population from state $|2\rangle$ and shelve it in the ground hyperfine state $|3\rangle$ of the ${}^3\text{H}_4$ manifold. This creates a very long-lived population pattern owing to the lifetime $T_1^g \approx 100 \text{ s}$ of the hyperfine ground states.

To measure the spatially varying population distribution after the BCP sequence, we apply a probe laser beam, with its frequency tuned to the $|1\rangle \leftrightarrow |2\rangle$ transition. We image the spatial beam profile of the probe laser after transmission through the crystal and compare it to a reference measurement with a fully transparent crystal. The probe beam propagates collinearly with the pump beam, but is temporally separated from the BCP sequences by approximately 7 ms. This delay significantly exceeds the excited state lifetime $T_1^e = 164 \mu\text{s}$, which ensures complete decay of any residual population in $|2\rangle$ before probing. The probe beam has a diameter of $550 \mu\text{m}$ (FWHM) in the crystal, which is significantly larger than the pump profile. Thus, the probe beam covers the full interaction region with the pump beam. We image the probe beam profile onto a CCD camera (Prosilica GC1290, Allied Vision) using an optical imaging system comprising lenses L2 (focal length 60 mm) and L3 (focal length 300 mm). A mechanical shutter [27] prevents the camera from being saturated by the pump beam.

To characterize the imaging system, we determined its magnification and resolution using a USAF-1951 target mask placed in the beamline instead of the crystal. Accounting for the additional crystal–vacuum interface with a refractive index of $n = 1.8$, the measured resolution is $5.4 \mu\text{m}$. This is only marginally larger than the diffraction limit at the numerical aperture $\text{NA} \approx 0.12$ of the imaging system. However, because of the non-negligible thickness of our 1 mm crystal, the small depth of field yields an actual resolution limit of approxi-

mately $10 \mu\text{m}$. The measured magnification of 4.97 matches the ratio of the focal lengths of lenses L2 and L3.

The optical transition in Pr:YSO is inhomogeneously broadened to several GHz, while the hyperfine splittings are on the order of 10 MHz. As a result, a single-frequency laser simultaneously couples all nine transitions among the six hyperfine states of the ${}^3\text{H}_4 \leftrightarrow {}^1\text{D}_2$ transition in Pr:YSO [see Fig. 1(a)] for ions in different frequency ensembles within the inhomogeneous line. To prepare the required level scheme and population distribution, we apply an optical pumping sequence. The preparation beam counterpropagates with the pump (and probe) beam at a small angle of approximately 2° . Collimated by lenses L4 (focal length 75 mm) and L2, the preparation beam has a diameter of $600 \mu\text{m}$ (FWHM), ensuring good overlap with the pump beam across the crystal length. The preparation sequence generates a “spectral pit”, i.e., a broad spectral hole with negligible absorption, which covers the pump and probe transitions, followed by a repump pulse that generates a spectral antihole on the pump transition with a residual linewidth of about $\Gamma = 2\pi \times 100 \text{ kHz}$ (FWHM). Finally, a cleaning pulse removes residual population from state $|3\rangle$, establishing the required population distribution. More details on the preparation sequence can be found in [28,29].

IV. EXPERIMENTAL RESULTS

We now present and discuss our experimental results on localization of atomic excitation driven by several classes of BCP sequences in Pr:YSO. For the composite sequences, we employ pump pulses with Gaussian temporal shape of duration $\tau = 230 \text{ ns}$ (FWHM) and a peak Rabi frequency of $\Omega_p = 2\pi \times 1.2 \text{ MHz}$. The pulses are truncated to a total duration of $t_p = 1 \mu\text{s}$ and separated by a short pause of $\Delta t = 40 \text{ ns}$. This delay serves to avoid phase errors, which otherwise would occur because of the finite rise time of the acousto-optic modulators.

Under these conditions, the total duration of the longest composite sequence we consider (with $N = 25$ pulses) remains below $t_C \lesssim 30 \mu\text{s}$, which is well within the coherence time $T_2^e = 111 \mu\text{s}$ of the optical transition $|1\rangle \leftrightarrow |2\rangle$ and is also well below the decay time $T_1^e = 164 \mu\text{s}$. This ensures that decay into states $|1\rangle$ and $|2\rangle$ is negligible before we apply a temporally rectangular shelving π pulse on the transition $|2\rangle \leftrightarrow |3\rangle$ with peak Rabi frequency $\Omega_{\text{shelve}} = 2\pi \times 0.35 \text{ MHz}$ and duration $t_{\text{shelve}} = 1.43 \mu\text{s}$. Thus, all Rabi frequencies remain significantly below the hyperfine splitting of the excited states ($\approx 4.5 \text{ MHz}$), allowing us to neglect couplings to additional states that could otherwise alter the population transfer dynamics.

To enable comparison of different BCP sequences, we keep the pulse parameters fixed and vary only the number of pulses and their phases. When using pulse areas other than $A = \pi$, we keep the laser intensity constant and vary the duration τ accordingly. For each BCP sequence, the population P_1 is measured on transition $|1\rangle \leftrightarrow |2\rangle$ using a probe pulse with a Gaussian temporal intensity profile of duration $10 \mu\text{s}$ (FWHM) and a peak Rabi frequency of approximately $\Omega_{\text{probe}} = 2\pi \times 15 \text{ kHz}$. This configuration ensures negligible perturbation of the population distribution.

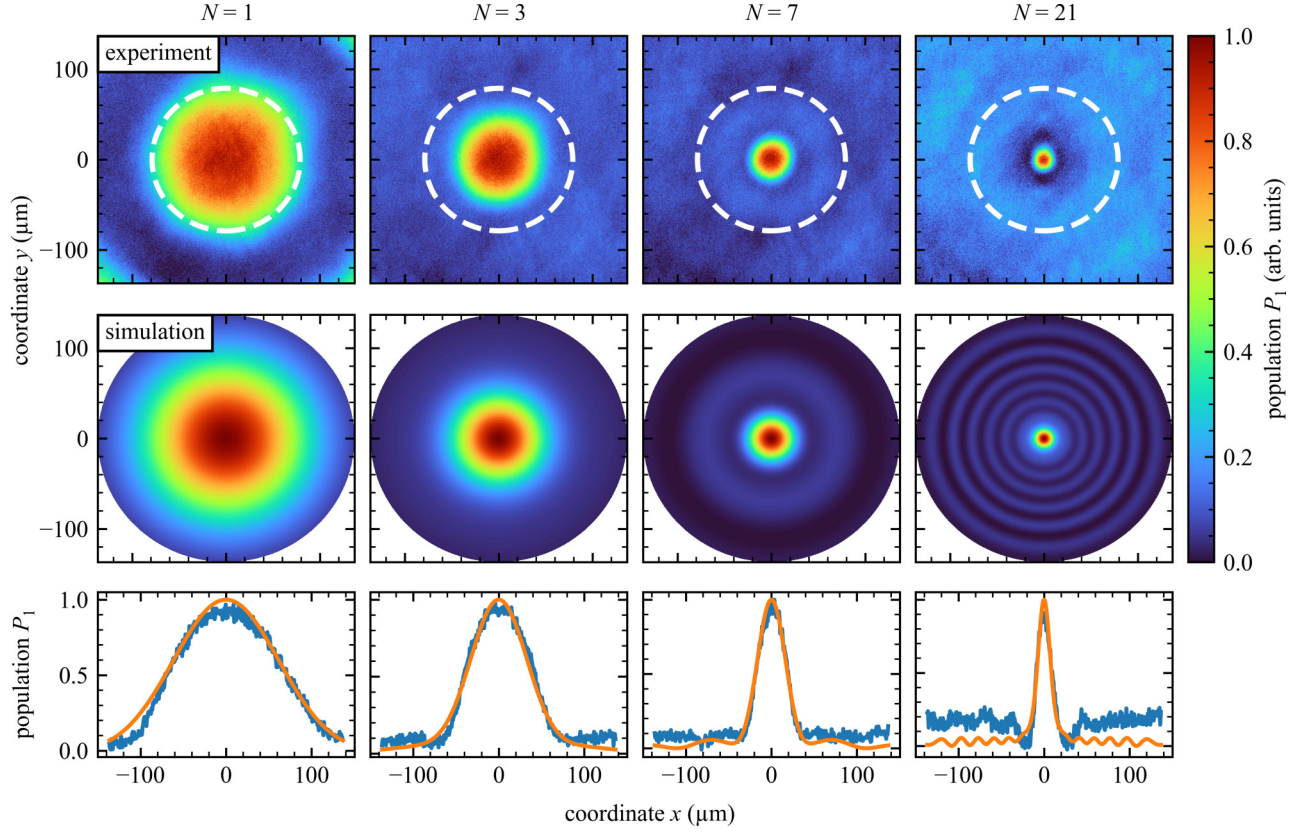


FIG. 4. Localization of atomic excitations in Pr:YSO by IHO BCP sequences. Variation of the population $P_1(x, y)$ vs spatial coordinates x and y across the pump beam profile. (Upper row) Experimental data for different numbers of pulses N . The white dashed circles indicate the pump beam waist. (Middle row) Numerical simulations. (Lower row) Horizontal cuts through the experimental data (blue line) and simulation (orange line) at coordinate $y = 0$.

A. Spatially confined excitations by IHO BCP sequences

We now investigate the features and performance of our new IHO BCP sequences. The top row of Fig. 4 shows the experimentally determined spatial population distribution P_1 after IHO BCPs with different number of pulses $N \leq 21$. Let us first consider the case of conventional excitation with a single π pulse ($N = 1$). In this case, the diameter of the measured population pattern is about $120 \mu\text{m}$ (FWHM), which we will call the π -pulse excitation width to use as a reference to benchmark the localization capabilities of BCP sequences. This turns out to be quite similar (although not equal due to different size measures) to the laser beam waist of $w_p = 160 \mu\text{m}$ ($1/e$ width), indicated in the figure by the white dashed circles. The population distribution shrinks considerably, when we apply composite pulses. Even with the shortest BCP sequence of $N = 3$ pulses, the improvement is already significant, reaching a confinement of 57% compared to the width achieved with a π pulse. The more pulses in the composite sequence, the stronger the spatial confinement as the robustness improves, finally reaching 11% of the π -pulse excitation width for a BCP sequence of $N = 21$ pulses. We note that Rabi oscillations (which are visible at the edge of the data set for the single π pulse and one might naively also expect such oscillations for any sequence of 3π pulses) are suppressed in the BCP-driven dynamics.

We compare the experimental data to a numerical simulation (see middle row of Fig. 4), which takes population decay, decoherence, as well as inhomogeneous broadening on the optical transitions $|1\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ into account. For the simulation, we assume a perfect TEM01* doughnut spatial mode for the pump Rabi frequency distribution and otherwise set all pulse parameters to the experimental values. The simulation is in excellent agreement with the experimental data. There are some minor deviations in the central peak of the population distribution, in particular for the single pulse $N = 1$, which is probably caused by residual deviations of the pump beam profile from a pure TEM01* mode. Nevertheless, the data and simulations for the BCP sequences clearly confirm that the population in a sharp region around the dark center of the doughnut beam remains unaffected—and that this region shrinks with increasing length of the composite sequences. We note that the simulations also show some residual, fast oscillations in the far wings of the excitation profile (see, e.g., the simulation and data for $N = 21$ pulses in Fig. 4). However, the oscillations are of very small amplitude only. In the experiment, our imaging system was not able to resolve them spatially and averages the oscillations to a low, constant background (see, e.g., cut through the data for $N = 21$ in the lower row of Fig. 4). This also merges with some incoherent background, which probably stems from

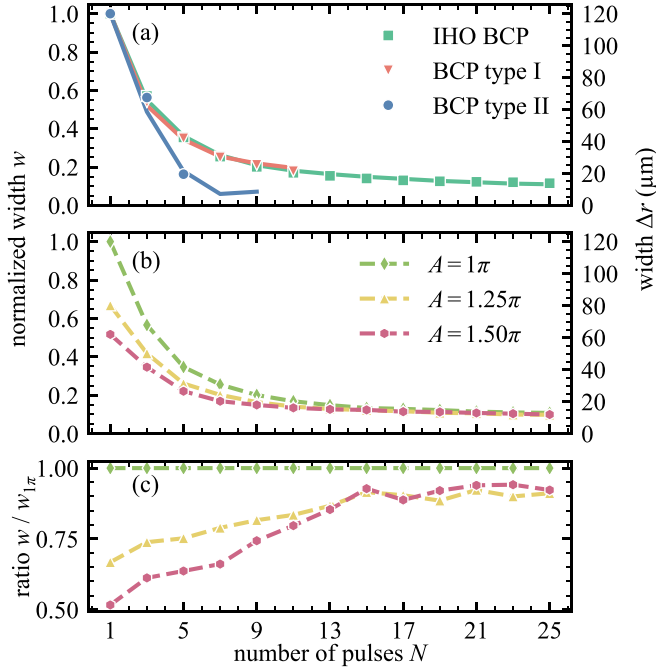


FIG. 5. (a) Normalized width w relative to the excitation width obtained with a single π pulse and actual width Δr (FWHM) of the population distribution $P_1(x, y)$ after localization by different BCP sequences vs number of pulses N . Comparison of experimental data (symbols) and numerical simulations (lines). (b) Width of the population distribution $P_1(x, y)$ after IHO BCPs with different peak pulse areas $A = \Omega_p \cdot \tau$ vs number of pulses N . (c) Same as (b), but localization width w now given relative to the measured width for π pulses. The pump beam had a waist of $w_p = 160 \mu\text{m}$.

decay after either off-resonant coupling to other excited states or from Raman scattering. We confirmed that this background is proportional to the pump beam energy and similar to previous investigations on NCP sequences in Pr:YSO [25]. As we apply a doughnut-shaped pump beam in the BCP pulses, the incoherent background contributes mainly to signal outside the central region. Despite our efforts to fully suppress the perturbing, ring-like excitation structures by the IHO BCP sequences (compare Fig. 2), there are still faint rings visible in the simulation, especially for $N = 7$. In the experiment, these rings vanish almost fully in the aforementioned background.

For a more thorough analysis of the achieved spatial confinement with the number of pulses N in the BCP sequences, we draw the attention of the reader now to Fig. 5. Plot (a) in this figure shows the widths of the experimentally determined population patterns after IHO BCP sequences (green squares) and the numerical simulation (green line) for up to $N = 25$ pulses normalized to the π -pulse excitation width. The simulation fits very well with the experimental data. As expected, the extension of the region with population confined in state $|1\rangle$ shrinks monotonously with the number of pulses N in the BCP sequence down to a localization of $14 \mu\text{m}$ for $N = 25$ pulses, i.e., around 11% of the value obtained for excitation with a single π pulse ($N = 1$). This clearly confirms the strong performance of BCP sequences and is already much better than the best results obtained for NCP sequences in previous experiments (25% at $N = 31$ for antisymmetric NCP

TABLE I. Convergence rates α for different BCP sequences, following fits of experimental data and numerical simulations with Eq. (1).

Sequence type	Convergence rate α	
	Experiment	Simulation
IHO BCP	0.68	0.67
BCP type I	0.67	0.67
BCP type II	0.95	0.96

sequences [25]), which in turn was already much better than what was achieved with other composite pulses [17].

The variation of the measured width Δr with the number of pulses N can be fitted, as theoretically deduced in Appendix B of [25], by a power function

$$\Delta r(N) = N^{-\alpha}, \quad (1)$$

with the convergence rate α . The latter quantifies the performance of composite sequences for localization of optical excitations. The larger α , the faster the convergence to strong spatial confinement. From the results in Fig. 5(a), we obtain exponents $\alpha_{\text{exp}} = 0.68$ for the experimental data and $\alpha_{\text{theo}} = 0.67$ for the numerical simulation, i.e., almost perfect agreement. The obtained convergence rate is much better than the value from the best antisymmetric NCP sequences in previous experiments, which yielded only $\alpha_{\text{exp}} = 0.44$ and $\alpha_{\text{theo}} = 0.48$.

B. Comparison with other BCP sequences

We will now experimentally compare the performance of our new IHO BCP sequences to the theoretically proposed BCP type I and II, which were never experimentally tested nor applied to spatially confine atomic excitations before. BCP type I sequences consist of pulses with an equal pulse area $A = \pi$, as we also apply them in IHO BCPs. On the other hand, BCP type II sequences consist of pulses with varying pulse area, taken as a free control parameter in addition to their phases. The theoretical work in [24] determines several subclasses of the two types with different error values ϵ , which defines the maximum allowed infidelity for which the transition probability remains above $P_2 = 1 - \epsilon$ within the numerical optimization process. For localization of atomic excitations, it is most suitable to choose the subclass with largest ϵ . This allows for the largest robustness vs variations of the pulse area and, thus, for the strongest spatial confinement—at the expense of a small drop in fidelity. We note that for the IHO BCP sequences in the present work, we also derived longer sequences up to $N = 25$, well beyond the maximum of $N = 11$ given for the BCP type I sequences in [24].

We now compare the performance of BCP type I and II with IHO BCPs [see Fig. 5(a)]. The experimental data for all sequences show excellent agreement with the corresponding numerical simulations. We fitted the variation of the localization width vs the pulse number via Eq. (1). Table I lists the convergence rates α . We find that the performance of the IHO BCPs and the BCP type I sequences is similar—which is not surprising as the numerical procedure of their derivation is similar (see below and Appendix). However, we also find

a substantially larger convergence rate for the BCP type II sequences. With only $N = 5$ pulses, they already show similar performance to the IHO BCPs with $N = 25$ pulses. We note that all BCP sequences outperform NCP sequences with their maximal convergence rate of $\alpha = 0.48$, as determined from theory and previous experiments [25].

The large convergence rate of the BCP type II sequences is because of the fact that the pulse area serves as one more control parameter (in addition to the relative phases) to optimize the population distribution. However, the obtained larger robustness vs variations of the pulse area comes at the costs of larger sensitivity to detuning variations [compare Fig. 2(b)]. In an inhomogeneously broadened medium such as Pr:YSO, this leads to perturbing ring-like population patterns around the central spot of spatially confined population (compare discussion in the theory section above). Because of the limited spatial resolution of our imaging setup in the experiment, the rings are not resolved and a cluttered background appears. For that reason, we could only obtain clear results in the experiment for BCP type II sequences with up to $N = 5$ pulses, and added simulations for $N = 7$ and 9 in Fig. 5(a). If the medium does not suffer from inhomogeneous broadening, a BCP type II sequence with $N = 7$ pulses could confine population down to 6% of the π -pulse excitation width. This is comparable to the performance of STIRAP-based localization [19], which may serve as a benchmark. However, as indicated above, the additional rings in the population pattern prepared by BCP type II sequences in inhomogeneously broadened media render them essentially not applicable in such systems. This is a clear advantage of the IHO BCP sequences.

Finally, we consider the robustness of BCPs with regard to variations of the intensity (pulse area), as already briefly discussed in the theory section above. Apart from the obvious benefit of robustness with regard to experimental fluctuations, e.g., when pulse area errors of the many pulses in the sequence arise, this also enables a further improvement towards even smaller localization widths. To do so, we increase the peak pulse area in the maxima of the doughnut pump beam profile to values $A > \pi$ since, unlike NCP sequences, in BCPs we are not limited to $A = \pi$. If the composite sequence was sensitive to deviations of the pulse area from an exact π pulse, the larger pulse area would lead to back transfer and a significant drop in fidelity. However, there is no such problem for a IHO BCP. It is robust and, hence, also works efficiently if the pulse area varies from an exact π pulse. Even though there may be a drop in fidelity, a robust BCP sequence can compensate this to some degree. As the BCP sequence permits us to increase the peak pulse area in the maxima of the doughnut beam profile, we also get larger pulse area close to the node of the profile. This leads to improved transfer in the adjacent region around the node and, thus, stronger spatial confinement of population P_1 .

We performed a systematic investigation of our IHO BCP sequences for different peak pulse areas of $A = 1\pi$, 1.25π , and 1.50π [see Fig. 5(b)]. The data show the decrease of the localization width (increasing spatial confinement) with increasing pulse count N , for different pulse areas in the IHO BCP sequence. We find that the localization indeed improves, but the effect is most pronounced at low pulse count, where there still is significant back transfer, and the advantage van-

ishes for higher N . This is especially apparent if we plot the ratio of the localization widths w for a BCP with pulse areas $A > \pi$ vs the localization width obtained for a sequence with exact π pulses [see Fig. 5(c)]. As we see from that experimental data, larger peak pulse areas permit stronger spatial confinement, i.e., smaller localization width—as expected for the robust BCP sequences. We should note, however, that improved localization by larger peak pulse area comes at the cost of reduced fidelity of the transfer process. Only for sequences with $N \geq 5$, the robustness of the sequences is sufficiently strong to suppress excitation in the wings of the beam profile. Nevertheless, even at $N = 5$ and a peak pulse area of 1.25π , we can further improve the localization width by 25% and for a pulse area of 1.50π even by 35% compared to π pulses. Depending on the application, this stronger confinement might be valuable even at reduced fidelity.

V. CONCLUSIONS

We experimentally demonstrated strong spatial confinement of optical excitations by broadband composite pulse (BCP) sequences in a rare-earth ion doped crystal (Pr:YSO). To this end, we developed novel BCP sequences, which are specifically optimized to compensate for variations of detunings, e.g., as relevant for applications in inhomogeneously broadened media such as Pr:YSO. We experimentally investigated the performance of the BCP sequence and compared it with numerical simulations, which agree very well with the experimental data. As an interesting feature, the BCP sequence is also robust with regard to fluctuations in the driving laser intensity. This provides experimental stability and enables further improvements to localize optical excitations by choosing pulse areas $A > \pi$ in the pulses of the sequence. We demonstrated that our BCP sequence spatially confined optical excitations in Pr:YSO to about 11% of the π -pulse excitation width, i.e., also very far below the beam waist of the driving doughnut-shaped laser beam. This is an improvement by over a factor of 2 when compared to narrowband composite pulses (NCP), applied in previous work. Moreover, we compared the IHO BCP to other composite pulses for broadband excitation (without specific optimization for inhomogeneously broadened media), which were theoretically developed in previous work but never tested in the experiment before. The latter include pulse sequences which use the pulse area as an additional control parameter. These sequences permit even stronger localization to approximately 6% of the width achieved with a single π pulse. However, the large robustness in pulse area comes at the expense of higher sensitivity in detuning, which leads to perturbations in the population pattern outside the central peak of strongly confined excitation. We also confirmed these findings by numerical simulations. While the spatial resolution of our experiment was limited by the imaging system to roughly 10 μm , the concept of BCPs is applicable also to spatially confine excitations below the diffraction limit of the laser beam. This will offer attractive options for high-resolution optical imaging, preparation, or addressing based on composite pulses, with applications in quantum technologies and classical applied optics.

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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: DERIVATION OF IHO BCP SEQUENCES

In order to derive our new BCP sequences, we consider a two-state quantum system interacting with an external coherent field. The Schrödinger equation

$$i\hbar\partial_t\mathbf{c}(t) = \mathbf{H}(t)\mathbf{c}(t) \quad (\text{A1})$$

describes the evolution of the qubit, where $\mathbf{c}(t) = [c_1(t), c_2(t)]^T$ is a column vector with the probability amplitudes of the two states $|\psi_1\rangle$ and $|\psi_2\rangle$. The Hamiltonian after the rotating-wave approximation is

$$\mathbf{H}(t) = (\hbar/2)\Omega(t)e^{-iD(t)}|\psi_1\rangle\langle\psi_2| + \text{H.c.}, \quad (\text{A2})$$

with $D = \int_0^t \Delta(t')dt'$, where $\Delta = \omega_0 - \omega$ is the detuning between the field frequency ω and the Bohr transition frequency ω_0 . For a constant detuning, we have $D = \Delta \cdot t$. The Rabi frequency $\Omega(t)$ is a measure of the field-system interaction energy $\hbar\Omega(t)$. We describe the evolution of the quantum system by means of the propagator $\mathbf{U}(t_f, t_i)$, which connects the probability amplitudes at the initial time t_i and the final time t_f : $\mathbf{c}(t) = \mathbf{U}(t, t_i)\mathbf{c}(t_i)$. For the sake of simplicity, hereafter we drop the temporal arguments in $\mathbf{U}$.

Because the 2×2 propagator is unitary, we can parametrize it by two complex Cayley-Klein parameters a and b as

$$\mathbf{U} = \begin{bmatrix} a & b \\ -b^* & a^* \end{bmatrix}. \quad (\text{A3})$$

For resonant excitation ($\Delta = 0$) with a pulse area A , we have $a = \cos(A/2)$ and $b = -i\sin(A/2)$. For an off-resonant pulse of rectangular temporal shape we have

$$a = \cos\left(\frac{1}{2}\sqrt{\Delta^2 + \Omega^2}T\right) + \frac{i\Delta \sin\left(\frac{1}{2}\sqrt{\Delta^2 + \Omega^2}T\right)}{\sqrt{\Delta^2 + \Omega^2}}, \quad (\text{A4a})$$

$$b = -\frac{i\Omega \sin\left(\frac{1}{2}\sqrt{\Delta^2 + \Omega^2}T\right)}{\sqrt{\Delta^2 + \Omega^2}}, \quad (\text{A4b})$$

where T is the pulse duration. There are very few other temporal shapes for which (more complicated) exact expressions for a and b are known (see, e.g., Refs. [30–33]); otherwise one has to compute a and b numerically.

A constant phase shift ϕ in the driving field, $\Omega(t) \rightarrow \Omega(t)e^{i\phi}$, is mapped onto the propagator as

$$\mathbf{U}(\phi) = \begin{bmatrix} a & be^{i\phi} \\ -b^*e^{-i\phi} & a^* \end{bmatrix}. \quad (\text{A5})$$

A sequence of N identical pulses—a CP sequence—each with a different pulse area A_k and a different phase ϕ_k ($k = 1, 2, \dots, N$),

$$(A_1)_{\phi_1}(A_2)_{\phi_2} \dots (A_N)_{\phi_N}, \quad (\text{A6})$$

produces the overall propagator (acting from right to left)

$$\mathbf{U}^{(N)} = \mathbf{U}(A_N, \phi_N) \dots \mathbf{U}(A_2, \phi_2)\mathbf{U}(A_1, \phi_1). \quad (\text{A7})$$

The transition probability is $P_2 = |U_{12}^{(N)}|^2$ and the no-transition probability is $P_1 = |U_{11}^{(N)}|^2$. Complete population inversion means $P_1 = 0$ and $P_2 = 1$.

If the pulse areas A_k and the phases ϕ_k are chosen appropriately one can modify the excitation profile essentially in any desired manner. In particular, one can nullify the first few terms in the Taylor expansion of the transition probability P_1 against a certain parameter at a specific value. Thereby, the composite transition probability can be made much more robust to variations in this parameter in the vicinity of the chosen value than the single-pulse transition probability. CPs can be made robust to variations in essentially any desired experimental parameter as well as in several parameters simultaneously. Moreover, the transition probability can be locked to 1, or 0, or any other value in between. In this manner, one can obtain broadband (with flat top), narrowband (with flat bottom), and passband (with both flat top and flat bottom) excitation profiles.

In this paper, we are interested in *broadband* CPs, which produce a very broad transition probability profile centered at the pulse area of π , which quickly decreases to 0 at pulse areas 0 and 2π .

The CPs derived by cancellation of the first few coefficients in the Taylor expansion of the transition probability at the selected parameter value (which is $A = \pi$ for BCPs) produce excitation profiles with wings of extremely high value within a certain range of $A = \pi$. Such extremely high values are not necessary in the present context. Instead, for the ultra-broadband pulses [24] the central part of the excitation profile is maintained above a certain high threshold value $1 - \epsilon$. Then one can obtain CPs with broader intervals of the pulse area wherein the error ϵ remains below a selected threshold. The larger the admissible error ϵ , the broader this interval can be. This results in a further broadened (ultra-broadband) excitation profile compared to the standard broadband pulses (obtained by canceling derivatives).

The BCP sequences of Ref. [24] have been obtained by numerically maximizing the value A_ϵ , for which the no-transition probability remains below the threshold ϵ within the pulse area range $A \in [A_\epsilon, 2\pi - A_\epsilon]$. Maximizing A_ϵ implies broadening the excitation profile toward $A = 0$ and 2π and enhancing the broadband effect. These sequences have been derived under the condition of exact resonance, $\Delta = 0$, while ignoring the excitation probability for $\Delta \neq 0$.

However, in inhomogeneously broadened ensembles of two-state atoms with transition frequencies within a certain

TABLE II. Phases (in units of π) of IHO BCP sequences. The sequences are symmetric, i.e., of the form $(\phi_1, \phi_2, \dots, \phi_{l+1}, \dots, \phi_2, \phi_1)$. Therefore, we list only the first $l + 1$ phases here.

Pulse count N	Phases $(\phi_1, \phi_2, \dots, \phi_{l+1})$
3	(0, 0.5991)
5	(0, 0.6658, 0.4102)
7	(0, 0.6829, 0.5535, 0.8623)
9	(0, 0.6912, 0.6116, 0.945, 0.776)
11	(0, 0.6959, 0.6406, 0.9886, 0.88, 1.0729)
13	(0, 0.6987, 0.6575, 1.0144, 0.9381, 1.1465, 1.0172)
15	(0, 0.7005, 0.6684, 1.031, 0.9748, 1.193, 1.0996, 1.2430)
17	(0, 0.7033, 0.6806, 1.051, 1.0129, 1.2418, 1.1752, 1.3304, 1.2253)
19	(0, 0.7041, 0.6858, 1.0586, 1.0305, 1.2642, 1.2145, 1.377, 1.2982, 1.4132)
21	(0, 0.705, 0.6907, 1.0662, 1.0465, 1.2848, 1.2486, 1.4173, 1.3581, 1.4807, 1.3933)
23	(0, 0.7050, 0.6928, 1.0689, 1.0538, 1.2937, 1.2657, 1.4377, 1.3918, 1.5196, 1.4517, 1.5462)
25	(0, 0.7221, 0.6826, 1.0980, 1.0479, 1.328, 1.2753, 1.4783, 1.4213, 1.5715, 1.505, 1.6166, 1.532)

bandwidth $[\omega - \Delta_0, \omega + \Delta_0]$ around the carrier frequency ω of the driving laser pulse, plenty of artifacts in the excitation probability may emerge within the detuning range $[-\Delta_0, \Delta_0]$ for pulse areas in the range $A \in [A_\epsilon, \pi]$, as shown in Fig. 2. Because the overall signal is an integral over this detuning range, the regions with appreciable probability within this range generate unwanted sidebands around the central peak in the experiment.

Therefore, we have developed new inhomogeneous-optimized ultra-broadband CPs, which clean up the high-probability artifacts within the detuning range $[-\Delta_0, \Delta_0]$ and the pulse area range $A \in [A_\epsilon, \pi]$. We have done this by numerically minimizing the value A_ϵ , for which the no-transition probability remains below the value ϵ within the pulse area range $A \in [A_\epsilon, \pi]$ and the detuning range $[-\Delta_0, \Delta_0]$. In so doing, we have assumed rectangular pulse shapes.

This numerical procedure has suggested that (i) the constituent pulses should be an odd number, $N = 2l + 1$, (ii) all pulse areas in the composite sequence should be equal, and (iii) the composite phases should be symmetric. The resulting composite sequences read

$$A_{\phi_1} A_{\phi_2} \cdots A_{\phi_l} A_{\phi_{l+1}} A_{\phi_l} \cdots A_{\phi_2} A_{\phi_1}. \quad (\text{A8})$$

This choice ensures the transition probability $P_2 = 1$ for $A = \pi$. The transition probability is invariant to a constant phase shift in all phases. Therefore, we can set the first and last phases to zero. We are then left with a maximization problem for A_ϵ with l free parameters, namely the phases from ϕ_2 to

ϕ_{l+1} . For $\epsilon = 1\%$, this gives us the IHO sequences discussed in the main text. We list the phases of these sequences in Table II. We note that these sequences are not unique. There are multiple solutions to the maximization problem yielding the same excitation probability, because of the periodic structure of the involved functions.

Specifically, the derivation of the ultra-broadband composite sequences follows the procedure described below:

(1) Use the symmetric sequence (A8) for a specific number of pulses.

(2) Fix the detuning range $[-\Delta_0, \Delta_0]$ using feedback for the inhomogeneous broadening from the experiment; for the present purposes, we use $\Delta_0 = 0.05/T$.

(3) Select a sufficiently small value A_ϵ and start with the corresponding phases of Ref. [24].

(4) Find (via random search and gradient descent) a set of phases in a small vicinity of the initial phases such that the transition probability P_2 remains above the value $1 - \epsilon$ throughout the box defined by $\Delta \in [-\Delta_0, \Delta_0]$ and $A \in [A_\epsilon, \pi/2]$.

(5) If the search in (4) succeeds then use the phases found in it as starting values, increase the value of A_ϵ and repeat (4).

(6) Repeat the loop of (4) and (5) until no set of phases is found that satisfies the condition in (4). The last set of phases satisfying this condition and corresponding to the largest possible value of A_ϵ is retained as the optimal set for the particular pulse sequence (A8) and listed in Table II.

Table II lists the phases of the IHO BCP pulse sequences, developed and demonstrated in this work. The parameters of the known sequences with equal and varying pulse areas are available from literature [24].

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