Spatial and spectral coherent control with frequency combs

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Quantum coherent control\textsuperscript{1-6} is a powerful tool for steering the outcome of quantum processes towards a desired final state by the accurate manipulation of quantum interference between multiple pathways. Although coherent control techniques have found applications in many fields of science\textsuperscript{7-12}, the possibilities for spatial and high-resolution frequency control have remained limited. Here, we show that the use of counter-propagating broadband pulses enables the generation of fully controlled spatial excitation patterns. This spatial control approach also provides decoherence reduction, which allows the use of the high-frequency resolution of an optical frequency comb\textsuperscript{13,14}. We exploit the counter-propagating geometry to perform spatially selective excitation of individual species in a multicomponent gas mixture, as well as frequency determination of hyperfine constants of atomic rubidium with unprecedented accuracy. The combination of spectral and spatial coherent control adds a new dimension to coherent control, with applications in nonlinear spectroscopy, microscopy and coherent control adds a new dimension to coherent control, with frequency combs

where $A(\omega)$ is the spectral amplitude and $\Phi(\omega)$ is the spectral phase at a frequency $\omega$ of the laser field, relative to half the transition frequency ($\omega_0/2$). The appearance of the fields $A(\omega)$ and $A(-\omega)$ in the integral reflects the fact that many pairs of different spectral modes centred around $\omega_0/2$ are involved in the two-photon transition.

Some distinct properties of the excitation profile can be derived even without full evaluation of this integral. In the case of an antisymmetric phase function $\Phi(\omega) = -\Phi(-\omega)$, the phase-dependent terms in the exponent of each integral cancel. The excitation probability will therefore be identical to the case of transform-limited pulses, which is a behaviour similar to that observed in the single beam configuration\textsuperscript{17}. A new level of control is made possible by applying symmetric spectral phase masks, which results in the generation of complex spatial patterns. Remarkably, when equation (1) is integrated over the spatial dimension (simulating a position-insensitive detector), all phase terms drop out, resulting in optimal constructive interference. The spatial excitation pattern can thus be manipulated without affecting the total signal strength.

As a proof-of-principle we conducted measurements on various two-photon transitions in atomic rubidium and caesium (level diagrams are presented in Fig. 1a). The experimental set-up is shown in Fig. 1b. Briefly, femtosecond pulses from a frequency comb laser are spectrally shaped in a standard phase-only 2F-2F configuration. The shaped pulses are focused in a vapour cell containing either pure rubidium or a rubidium/caesium mixture. After passing the cell, the pulses are reflected back at a distance that matches the repetition frequency of the laser, so that every two consecutive pulses overlap in the middle of the vapour cell. Control over the atomic excitation is achieved by applying a spectral phase mask to the pulse shaper and tuning the frequency comb parameters $f_{\text{rep}}$ (repetition frequency) and $f_{\text{c}}$ (carrier-envelope offset frequency), which determine the absolute frequencies of the two-photon comb modes through the comb equation $f_n = f_{\text{c}} + n \times f_{\text{rep}}$ (refs 13,14), where $n$ is an integer. The excited state population is monitored by detecting the ultrashort pulses. Atoms at different positions experience the two counter-propagating pulses at different times with a delay $\Delta t = 2z/c$ ($z = 0$ is the point where the unshaped pulses overlap and $c$ is the speed of light). The excitation probability from counter-propagating pulses can be written as follows (for a complete derivation see Supplementary Section S1):

\begin{equation}
S_{\omega}(\omega) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} A(\omega')A(-\omega')A(\omega')A(-\omega')d\omega'd\omega
\end{equation}

where $A(\omega)$ is the spectral amplitude and $\Phi(\omega)$ is the spectral phase at a frequency $\omega$ of the laser field, relative to half the transition frequency ($\omega_0/2$). The appearance of the fields $A(\omega)$ and $A(-\omega)$ in the integral reflects the fact that many pairs of different spectral modes centred around $\omega_0/2$ are involved in the two-photon transition.

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The localized signal in the centre results from the spatial overlap of two counter-propagating transform-limited pulses.

A harmonic spectral phase mask alters the temporal intensity distribution of the pulses, breaking each pulse into a train of sub-pulses. In Fig. 2b, a sine-modulated spectral phase ($\varphi = \pi/2$, antisymmetric relative to $\omega_b/2$) is applied with a modulation depth of $\alpha = 1.2$. An antisymmetric spectral phase does not influence the single-sided excitation\(^\text{17}\), which appears in our case as an unchanged, spatially independent background. We observe that the localized signal resulting from counter-propagating pulses is also unaffected by an antisymmetric phase, as predicted from equation (1). Although the sub-pulses overlap in multiple regions in space, the quantum interference is only constructive in the centre.

Figure 2c shows an image of the excitation pattern when applying a symmetric phase mask ($\varphi = 0$) with the same modulation depth and modulation frequency as in Fig. 2b. Even though the temporal intensity distributions of the interacting pulses for Fig. 2b and c are identical, the resulting spatial excitation patterns differ substantially. For this symmetric phase mask, the single-sided excitation is completely eliminated, which is why such pulses have been named 'dark pulses'\(^\text{17}\). However, Fig. 2c shows that counter-propagating dark pulses actually give rise to a spatially structured two-photon excitation pattern where the sub-pulses overlap. We find that these counter-propagating dark pulses are not only not dark, but induce the same total excitation as transform-limited pulses.

Variation of the modulation depth can be used as a ‘switch’ to turn the single-sided signal on and off, but can also be used to control the relative brightness of the individual excitation regions. In Fig. 2d, the modulation depth was set to $\alpha = 2.76$, where the single-sided signal is again eliminated completely. The fluorescence image displays additional excitation regions with different ratios between their relative intensities. In addition, the positions of the excitation regions can be controlled by varying the modulation frequency $\beta$ (Supplementary Section S2).

In the second set of measurements we illustrate the versatility of spatial coherent control by spatially separating the excitation of different atomic species. We used a rubidium/caesium mixture vapour cell and tuned the comb parameters to $f_0 = 25$ MHz and $f_{\text{ref}} = 178,435,286$ Hz, for which both the $^{85}\text{Rb}$ $5S(F = 3) \rightarrow 7S(F = 3)$ transition in rubidium at 2 $\times$ 760 nm and the $^{133}\text{Cs}$ $6S(F = 4) \rightarrow 8S(F = 4)$ transition in caesium at $2 \times 822$ nm are simultaneously excited. Figure 3a shows the excitation profile with transform-limited pulses. The signal is dominated by single-sided excitation, and no distinction can be made between the atomic species. Subsequently, we used a V-shaped spectral phase mask ($\Phi(\omega) = \alpha(\omega - \omega_b/2)$) that effectively eliminated the single-sided signal. By applying such a spectral phase mask to both spectral regions (centred around $\lambda_{\text{Rb}} = 760$ nm and $\lambda_{\text{Cs}} = 822$ nm), we created a background-free excitation pattern as seen in Fig. 3b. By changing the slope of the V-shaped phase masks, the excitation positions of the individual species could be moved around independently. This resulted in controlled spatially separated excitation of rubidium and caesium atoms, as seen in Fig. 3c. We verified that excitation within the various regions in Fig. 3c originates from different atomic species by scanning the frequency comb modes over the different atomic transitions. Such a scan reveals the hyperfine atomic structure, which is a unique fingerprint of the different atoms (for details see Supplementary Section S3).

The combination of background-free signal and the reduction of Doppler-broadening\(^\text{18,19}\) is essential for performing high-precision direct frequency comb spectroscopy\(^\text{20-25}\). These requirements are
Figure 2 | Experimental demonstration of spatial coherent control using four different phase masks. a–d. Transform-limited pulses (a), antisymmetric sine-modulated spectral phase (\(\alpha = 1.2, \beta = 900\) fs, \(\varphi = \pi/2\)) (b), symmetric cosine-modulated spectral phase (\(\alpha = 1.2, \beta = 900\) fs, \(\varphi = 0\)) (c) and symmetric cosine-modulated spectral phase with a larger amplitude (\(\alpha = 2.76, \beta = 900\) fs, \(\varphi = 0\)) (d). For each figure we present the temporal intensity profile (depicted in red) of the counter-propagating pulses and the observed spatial excitation pattern. A cross-section of each fluorescence image is compared to a numerical evaluation of equation (1) (green line). The broadening of the data due to the imaging resolution and the atomic motion is taken into account in the theory by convolving all simulation curves with the same 80 \(\mu m\) full-width at half-maximum Lorentzian distribution. A correction for the non-uniform intensity due to the focusing geometry is derived by fitting the background with a single Gaussian distribution. The resulting correction (including a single scaling factor) is then applied to all theoretical curves.

Figure 3 | Spatial coherent control of different atomic species. a. Spatial excitation patterns using transform-limited pulses. b. Double V-shaped spectral phase mask (\(\Phi(\omega) = \alpha|\omega - \omega_0|/2\)) with \(\alpha = 0.5\) ps for both spectral regions. c. Double V-shaped spectral phase mask with \(\alpha = 0.5\) ps for caesium and \(\alpha = 1.1\) ps for rubidium. All images were taken with the same comb parameters.
The shaper system was based on a zero-dispersion 2f–2f configuration, with two 1.200 mm−2 gratings and two spherical mirrors with radii of curvature of 100 cm. A 640-pixel computer-controlled spatial light modulator (SLM) was placed at the Fourier plane to apply a phase-only spectral mask.

The excitation setup consisted of a vapour cell containing either pure rubidium or a rubidium/cesium mixture. The cell was heated to a temperature of 60–70 °C to increase the pressure in the cell for increased signal strength. The laser beams were focused in the middle of the cell. For spectroscopy measurements, the focus size was chosen to be larger than 50 μm to avoid transit-time broadening.

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Author contributions

K.S.E.E. conceived the V-shape phase concept for frequency comb spectroscopy. I.B. extended this concept to full spatial coherent control and provided the theoretical description. I.B. performed the experiments (with assistance from S.W and K.S.E.E.) and data analysis. K.S.E.E. supervised the project. All authors participated in the design of the experiments, interpretation of the results and writing of the manuscript.

Additional information

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Competing financial interests

The authors declare no competing financial interests.