Amplification of intense light fields by nearly free electrons

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Light can be used to modify and control properties of media, as in the case of electromagnetically induced transparency or, more recently, for the generation of slow light or bright coherent extreme ultraviolet and X-ray radiation. Particularly unusual states of matter can be created by light fields with strengths comparable to the Coulomb field that binds valence electrons in atoms, leading to nearly free electrons oscillating in the laser field and yet still loosely bound to the core1−3. These are known as Kramers–Henneberger (KH) states4, a specific example of laser-dressed states5. Here, we demonstrate that these states arise not only in isolated atoms4,5, but also in rare gases, at and above atmospheric pressure, where they can act as a gain medium during laser filamentation. Using shaped laser pulses, gain in these states is achieved within just a few cycles of the guided field. The corresponding lasing emission is a signature of population inversion in these states and of their stability against ionization. Our work demonstrates that these unusual states of neutral atoms can be exploited to create a general ultrafast gain mechanism during laser filamentation.

It is often assumed that photo-ionization happens faster in more intense fields. Yet, since the late 1980s, theorists have speculated that atomic states become more stable when the strength of the laser field substantially exceeds the Coulomb attraction to the ionic core1−4,6−11. The electron becomes nearly but not completely free: rapidly oscillating in the laser field, it still feels residual attraction to the core, which keeps it bound. The effective binding potential, averaged over the electron oscillations, is sketched in Fig. 1a. It has a characteristic double-well structure, the wells occur when the oscillating electron turns around near the core. The laser-modified potential also modifies the spectrum, with laser-induced shifts adding to the familiar ponderomotive shift associated with nearly free electron oscillations. We refer to these states as ‘strongly driven laser-dressed states’. In spite of many theoretical predictions, it took three decades before their existence was inferred in experiments2,4,5, showing neutral atoms surviving laser intensities as high as $I \approx 10^{15}–10^{16}$ W cm$^{-2}$. But are such unusual states really exotic? Can they also form in gases at ambient conditions, at intensities well below $10^{15}–10^{16}$ W cm$^{-2}$? After all, for excited electronic states bound by a few electronvolts, the laser field overpowers the Coulomb attraction to the core at $I \approx 10^{15}–10^{16}$ W cm$^{-2}$. If so, these states manifest inside laser filaments, the self-guiding light structures created by the nonlinear medium response at $I \approx 10^{14}$ W cm$^{-2}$ (ref. 15).

The formation of the Kramers–Henneberger (KH) states should be probed by a weak broadband (~5 fs) probe, carried at wavelength $\lambda = 600$ nm and centred in the middle of the pump pulse ($t = 0$). The time-dependent response to the probe, $\Delta d(t)$, is extracted from the full polarization $d(t) = \langle \Psi(t)|d|\Psi(t)\rangle$ as described in ref. 17: $\Delta d(t) = d(t) − d_0(t)$. Here $d_0(t) = \langle \Psi_0(t)|d|\Psi_0(t)\rangle$, where $d$ is the dipole operator in the acceleration form, and $\Psi(t)$ and $\Psi_0(t)$ are the time-dependent wavefunctions computed with both fields or the strong IR pump only, respectively.

The key quantity is $\text{Im}[\Delta D(o)]$, the imaginary part of the Fourier transform of $\Delta d(t)$: a negative imaginary part signifies gain, whereas a positive imaginary part signifies loss. Figure 1b,c shows a window Fourier transform of $\Delta d(t)$, using the sliding Gabor window.
G_{\text{KH}}(t,t_0) = \exp\left(-\frac{(t-t_0)^2}{T^2}\right) \text{ (where } T = 500 \text{ atomic units (a.u.)), which allows us to time-resolve the emission. Below } I = 10^{14} \text{ W cm}^{-2}, \text{ the time-dependent gain is offset by the loss, but the situation changes radically above this intensity: at } I = 1.4 \times 10^{14} \text{ W cm}^{-2} \text{ gain dominates and amplification lines arise around 550–570 nm and 630–650 nm (Fig. 1c, d). The lines are asymmetric, more Fano-like than Lorentzian (Fig. 1d), as expected in the presence of a strong driving field.}

Thus, theory predicts the emergence of gain at intensities \( I \approx 10^{14} \text{ W cm}^{-2} \), which will manifest in the forward spectrum from only shaped (that is, sharp rise time) laser pulses. Experimentally, we look for new, atypical absorption and emission structures with asymmetric Fano-like shapes, between 400 nm and 700 nm. Second, the population inversion should arise intra-pulse and depend on the pulse shape (rise time and duration). Third, the emission should have lasing characteristics and occur at transitions absent in the field-free atom or ion. To test these predictions we employ a pulse-shaping set-up with a Ti:Sapphire laser in combination with a 640-pixel spatial light modulator (SLM), providing 50 μJ pulses centred at 800 nm (Supplementary Fig. 1b, Methods). The pulses are focused into a chamber by a 300 mm off-axis spherical mirror, leading to a short filament (4 mm, see Supplementary Fig. 1a and Methods) in Ar or Kr (2–9 bar). The pulse is shaped such that it acquires the required sharp rise at the beginning of the filament, maximizing the population of the stabilized, strongly driven laser-dressed states. The pre-compensation of the desired pulse shape is achieved by acoustic shock wave optimization at the focus (see Methods). Pulse fronts of ~5 fs are generated, as measured using a spectral phase interferometry for direct electric field reconstruction (SPIDER).

Figures 2, 3 show the experimental results. The strongly driven laser-dressed states are best accessed using pulses with a sharp rise time. Thus we can compare the forward emission from pulses with the same spectra, but different temporal shapes. The red line in Fig. 2a shows the supercontinuum generated inside the filament, for a smooth, 40 fs, broad Gaussian laser pulse. This standard pulse yields a typical supercontinuum spectrum in the forward direction, with no resonant lines attributable to atoms or ions. In contrast, when the pulse rise is fast (that is, a 7 fs pulse), we observe dramatically different spectra with distinct asymmetric (Fano-like) amplification lines at 530 nm, 550 nm, 570 nm and 625 nm (Fig. 2a), as predicted by the theory. The Gaussian pulse has the seed radiation, laser-dressed states are best accessed using pulses with a sharp rise time.

Pulse-shaping control of gain is demonstrated when comparing an asymmetric triangular-like pulse (5 fs rise, 20 fs decay) against the reverse shape (20 fs rise, 5 fs decay). They have identical spectra but opposite spectral phase. The pulse with the fast rise generates strong gain lines, while the pulse with the slow rise leads to

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In Fig. 2c, the emission lines at 557 nm and 625 nm undergo absorption rise time, 10 fs plateau and 10 fs decay time, for increasing pulse energies. Two absorption features are visible at 625 nm and 560 nm, which become gain features at 33 µJ and 28 µJ, respectively, and a broad gain feature emerging at 600 nm. In both a and d, Forward emitted spectra, following filamentation in the same argon cell, of a 10 fs rise time, 5 fs plateau and 10 fs decay time, for increasing pulse energies. Gain features are visible at 625 nm, 650 nm and 670 nm. In both a and d, red arrows indicate the movement of emission peaks with increasing input energy, and the dashed lines are to guide the eye to specific lasing peaks. An input white light spectrum is shown in Supplementary Fig. 6.

The key role of the laser-dressed (KH) states is confirmed by the theoretical results in Fig. 3. We cross-check the shape and spectrum of the trapezoidal input pulse (10 fs rise, 10 fs plateau, 10 fs decay) at the onset of filament, using numerical pulse propagation simulations (see Methods). We then use the experimental pulse in the TDSE simulations to calculate the intensity of the emitted radiation. The simulated output spectrum is normalized to the input spectrum at the 800 nm carrier wavelength, as in the experiment. Figure 3b shows the emergence of strong emission lines, as in the experiment (Fig. 3a). Note these peaks emerge where Fig. 1d shows gain. Figure 3b also shows that the observed lines cannot be attributed to standard nonlinear effects during propagation: a simulation of laser filamentation using standard propagation models (see Methods) does not lead to any peaks in the spectral region of interest.

Finally, we focus on the spectral region between 610 nm and 690 nm. There are no field-free lines in the argon spectrum that coincide with the observed strong amplification lines at 625 nm and near 675 nm. However, Fig. 3c shows that transitions between the laser-dressed states (calculated in the KH frame, see Methods) do move into this region at \( I \approx 0.9 \times 10^{14} \text{ W cm}^{-2} \). Note that Fig. 3c
does not show the overall pondermotive shift of the excited states and demonstrates only the additional shift. This shift is small compared to the pondermotive shift, which reaches 6 eV at $10^{14}$ W cm$^{-2}$ (for $\lambda = 800$ nm). Figure 3d shows the population difference between the field-free states that move into this region at intensities around $10^{14}$ W cm$^{-2}$. These are the states with field-free transition frequencies between 500 nm and 600 nm, which acquire population inversion at intensities around $10^{14}$ W cm$^{-2}$.

The lasing mechanism is not specific to argon. Similar results were found in krypton (see Fig. 4 and Supplementary Fig. 7). The lasing transitions are at different energies than in argon, reflecting the different atom, but also exhibit both broad and narrow gain features and asymmetric Fano-like lineshapes.

There is no direct connection between the observed resonant widths of laser-dressed states, their lifetime and pulse duration. Indeed, the laser-dressed states undergo ultrastable dynamics intrapulse and their positions are intensity-dependent, leading to ‘inhomogeneous’ broadening due to the spatial and temporal intensity distributions. In a 7 fs pulse, the dressed states shift rapidly with changing pulse intensity, so that resonances should broaden with increasing peak intensity (Fig. 4a, from 3 nm to 7 nm at 617 nm). For a long ‘trapezoidal’ pulse (10 fs rise, 40 fs plateau, 10 fs decay), transition lines shift with intensity but keep their widths (~7 nm at 624 nm and ~12 nm at 613 nm; Fig. 4b).

The observation of gain lines specific to the atom dressed by an intense, $I > 10^{14}$ W cm$^{-2}$, laser field, and absent in the spectrum of field-free transitions, shows that the seemingly exotic KH states are ubiquitous even in dense (1–9 bar) gases interacting with strong laser fields. At high intensities, the laser-driven atom can become an inverted medium, inside the laser pulse, where electrons respond almost as free, yet remain bound and can be used as a multi-photon pumped gain medium during laser filamentation. Amplification at the inverted transitions between the dressed states, resulting in the emergence of gain lines during the pump pulse, can trigger additional wave-mixing processes with the strong pump, possibly leading to additional parametric gain lines in the spectrum. After the end of the pulse, coherent free induction decay can also seed lasing between the field-free states carrying population inversion. Our findings illustrate new opportunities for enhancing and controlling lasing inside laser filaments by optimizing the shape of the input laser pulse.

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**Fig. 3** | A comparison between simulated and experimental emission spectra from a 10 fs rise, 10 fs plateau, 10 fs decay laser pulse shape. a. Experimentally measured forward emission for filamentation in argon, at 50 µJ, showing the input (blue) and output (orange) spectra. b. Theoretically calculated emission spectrum of the strongly dressed atom for the experimental pulse at the input of the filament: input (blue), output (orange). The green line shows the results of filamentation propagation simulations (see Methods) without including the laser-dressed states. c. Position of the KH states as a function of laser intensity. To demonstrate the origin of the emission in the spectral region 650 nm ± 50 nm, the lowest excited KH state (blue) is shifted up (dashed and shaded blue): it enters the dense manifold of weakly bound states (grey region) at $I \approx 0.9 \times 10^{14}$ W cm$^{-2}$. d. Relative population difference between key field-free states, which can contribute to emission, between 500 nm and 700 nm, for two different intensities: $0.92 \times 10^{14}$ W cm$^{-2}$ (diamonds) and $1.37 \times 10^{14}$ W cm$^{-2}$ (circles). Dashed blue lines indicate that the population of the energetically lower state is higher than that of the energetically higher state. Orange lines indicate population inversion between the states. The population is calculated at the end of the pulse.
Fig. 4 | The forward emission spectra of trapezoid pulse shapes in krypton at 9 bar, with increasing pulse energy. a, A Fourier-limited pulse, 7 fs duration. A shift in the transition lines is observed with increasing pulse energy of 3–5 nm over 50 µJ, and at 20 µJ the weak emission/absorption lines are strongly enhanced and the linewidths broadened (from 3 nm to 7 nm). b, Trapezoid pulses with 10 fs rise time, 40 fs plateau and 10 fs decay time. Narrow and broad gain features are visible, experiencing a spectral shift of about 3–5 nm over an energy increase of 50 µJ, but with less broadening. A distinct Fano lineshape emerges at 627 nm. Dashed lines and white arrows are to guide the eye, between the figures, to show the shift in emission wavelengths between different pulse durations and also to highlight the field dependence of the states involved. The temporal pulse shape is shown in red for each graph.

Methods

Methods, including statements of data availability and any associated accession codes and references, are available at https://doi.org/10.1038/s41567-018-0105-0.

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Author contributions
J-PW and M.I. conceived the experiment. M.I., F.M., M.R., T.B. and O.S. performed the calculations and developed the theoretical interpretation. N.B. performed filamentation propagation simulations. M.M., S.H., J.K. and J.G. designed the experimental apparatus. A.P and A.L. designed and implemented the pulse-shaping process. M.M., A.P, A.I, J.G. and S.H. performed the experiment and pulse measurements. M.M., A.P and J.K. analysed and processed the experimental data. All authors contributed to the writing of the manuscript.

Competing interests
The authors declare no competing interests.

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Methods

Pulse-shape generation. To synthesize laser waveforms with pulse-shape control down to the few-cycle level, a CPA Ti:Sapphire laser, (780 nm, 1.5fL, 40 fs, 1 kHz; details and a diagram can be found in Supplementary Fig. 1b) undergoes two-stage filamentation in air, through loose focusing with 2 m and 1.25 m focal length mirrors. The pulse, broadened (700–900 nm) by the first filamentation stage, is re-collimated and recompressed with a pair of chirped mirrors before refocusing for the second filamentation stage, with a pair of spherical mirrors. At the exit of this second stage, the pulse spectrum spans more than one octave (450 nm–1 µm) and is recompressed by a chirp mirror arrangement. The final compression of higher spectral phase orders and the pulse shape control are achieved using a 4 f all-reflective pulse shaper with a dual mask, 640-pixel, liquid crystal modulator. In this configuration, few-cycle 5 fs pulses of up to 50 µJ can be produced, in addition to flat top, or sawtooth with sharp rise times. These are optimized using a pulse-shape optimization algorithm explained below.

Pulse-shape optimization and diagnostics. In order to compensate for dispersion arising from the chamber window and the propagation in the pressured gas before the focal point, we apply a phase detection algorithm, onto the SLM, to get the shortest pulse (Fourier transform-limited) at the focus. The signal used for the optimization loop was the acoustic shock wave released by the plasma, representative of the free-carrier density produced by the laser. Using subsequent measurements we verify this procedure leads to the desired pulse shape, at the onset of the filament (Ti: sapphire–limited, sawtooth, flat top taperspoids). The pulse shapes are measured using a transient-grating frequency-resolved optical grating (FROG), as well as a SPIDER (Ventcom), at pulse positions before and after filamentation. To measure the pulse shape within the filament, a 100 µm Al foil is placed in the filament path. The filament drills a self-adapted iris, arresting further filamentation and nonlinear propagation, but preserving the temporal pulse shape at this distance. The remaining beam was analysed by a SPIDER. The SPIDER traces are shown in Supplementary Figs. 4 and 5.

Pulse propagation simulations. Numerical simulations, based on a unidirectional laser pulse propagation equation (UPPE) 2, are used to simulate the laser filamentation process and cross-check the pulse-shape optimization routine described above. The propagation simulations are first carried out up to the onset of filamentation for sample pulses, and confirmed the desired experimental pulse shape. Next the same simulations were carried out throughout the full filamentation region to obtain the spectra both at the input and at the output of the filament. The numerical method and the code verification are described in detail elsewhere 24. Briefly, the simulations are performed in a cylindrically symmetric geometry, reducing the dimensionality of the problem to two spatial dimensions plus one temporal dimension. The ionization model uses the standard Perelomov–Popov–Terent’ev ionization rates. All standard nonlinear effects, such as self-focusing, self-phase modulation and self-steepening, are included (see Supplementary Fig. 8).

Filamentation in pressured argon and krypton cells. A schematic of the experimental set-up can be found in Supplementary Fig. 1. The shaped pulses enter a pressurized chamber (2–9 bar) containing Ar or Kr via 5 mm ultraviolet fused silica windows, where a 300 mm off-axis gold spherical mirror generates a filament 4–5 mm in length, before exiting the chamber through a 5 mm ultraviolet fused silica window. Spectra from the filament and its plasma are focused in the forward direction using a Gabor window in Fig. 1b,c, only the Gabor window was applied, without additional blackening. To obtain the dressed pulse shape at this distance. The remaining beam was analysed by a SPIDER. The SPIDER traces are shown in Supplementary Figs. 4 and 5.

Data availability. The data that supports the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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