Subcycle quantum electrodynamics

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Squeezed states1–4 of electromagnetic radiation have quantum fluctuations below those of the vacuum field. They offer a unique resource for quantum information systems5 and precision metrology6, including gravitational wave detectors, which require unprecedented sensitivity7. Since the first experiments on this non-classical form of light6,9, quantum analysis has been based on homodyning techniques and photon correlation measurements10,11. These methods currently function in the visible to near-infrared and microwave12 spectral ranges. They require a well-defined carrier frequency, and photons contained in a quantum state need to be absorbed or amplified. Quantum non-demolition experiments13,14 may be performed to avoid the influence of a measurement in one quadrature, but this procedure comes at the expense of increased uncertainty in another quadrature. Here we generate mid-infrared time-locked patterns of squeezed vacuum noise. After propagation through free space, the quantum fluctuations of the electric field are studied in the time domain using electro-optic sampling with few-femtosecond laser pulses15,16. We directly compare the local noise amplitude to that of bare (that is, unperturbed) vacuum. Our nonlinear approach operates off resonance and, unlike homodyning or photon correlation techniques, without absorption or amplification of the field that is investigated. We find subcycle intervals with noise levels that are substantially less than the amplitude of the vacuum field. As a consequence, there are enhanced fluctuations in adjacent time intervals, owing to Heisenberg’s uncertainty principle, which indicate generation of highly correlated quantum radiation. Together with efforts in the far infrared17,18, this work enables the study of elementary quantum dynamics of light and matter in an energy range at the boundary between vacuum and thermal background conditions.

In quantum electrodynamics, coherent states represent the closest counterparts to a classical electromagnetic wave. The quantum noise amplitudes of their electric and magnetic fields coincide precisely with those of the vacuum state19. Recently, the bare vacuum fluctuations of the mid-infrared (MIR) electric field were directly detected using highly sensitive electro-optic sampling based on ultrashort laser pulses15,16. One key aspect of this technique is that it operates in the time domain. Therefore, it should provide a resolution that is substantially below the duration of an oscillation period of any quantum field under study. It is tempting to consider an experiment that synchronously couples a non-classical state of light into the space-time volume that is probed, thus providing a quantum noise amplitude that deviates from pure vacuum fluctuations. An especially striking manifestation of quantum physics would be to demonstrate a spatial and temporal localization of less noise than that of the quantum vacuum. In conventional homodyning studies, the carrier wave of a local oscillator needs to be phase-locked to a quantum state11,16. In contrast, here we prepare a squeezed electromagnetic transient with a noise pattern that is synchronized with the intensity envelope of an ultrashort probe pulse. This tightly focused few-femtosecond optical wave packet then defines a subcycle space-time segment in which the quantum statistics of a MIR non-classical signal is sampled.

Our scheme to implement such an experiment is sketched in Fig. 1a. We send an intense near-infrared (NIR) pump pulse (red/yellow envelope) with a duration of 12 fs and centre frequency of 200 THz into a thin generation crystal (GX). In a first step, a carrier-envelope phase-locked electric field transient20 is generated by optical rectification (red line). Once built up, this transient starts to locally phase shift the co-propagating MIR vacuum fluctuations (green shaded band). This second step is achieved by means of the electro-optic effect in the GX, which establishes a change in refractive index $\Delta n(t)$ proportional to the MIR electric field amplitude $E_{\text{MIR}}(t)$. In a simplified picture, the resulting local anomalies in the speed of light might induce depletion of vacuum amplitude in certain space-time regions (blue shaded sections in Fig. 1a), piling it up in others (shaded in red). A high efficiency for this two-step mechanism to squeeze the MIR vacuum is ensured by the large second-order nonlinearity of the 16-μm-thick exfoliated piece of GaSe we use as the GX20. Tight focusing of the pump to a paraxial spot radius $w_{\text{pump}}$ of 3.6 μm also defines the transverse spatial mode for the non-classical electric field pattern. After the MIR field exits the GX, it is collimated and the residual pump is removed by a 70-μm-thick GaSb filter inserted at Brewster’s angle. A mode-matched 5.8-fs probe pulse (blue envelope in Fig. 1a) is then superimposed onto the MIR field and focused to $w_{\text{probe}}=3.6$ μm in a AgGaS2 detector crystal (DX) with a thickness of 24 μm (ref. 15). The probe pulse samples the electric field in the co-propagating space-time volume via the electro-optic effect15,20 and as a function of time delay $t_D$. We gain two different types of information: first, the coherent19 electric field amplitude $E_{\text{MIR}}(t_D)$ of the squeezing MIR transient is recorded in the conventional way20; and second, the quantum distribution of the MIR electric field is accessed via statistical readout15. Our technique allows us to directly reference the local noise level $\Delta E_{\text{rms}}$ in a squeezed transient (blue and red distributions in Fig. 1b) to the fluctuations $\Delta E_{\text{vac}}$ obtained under bare vacuum input (green distribution in Fig. 1b; see Methods for details). Relative differential noise (RDN) patterns mirroring $\Delta E_{\text{rms}}$ are then recorded as a function of delay time $t_D$ (see Fig. 1c). Only 4% of the total fluctuation amplitude in our set-up results from a bare MIR vacuum input; the rest is due to the noise-equivalent field of the detector $\Delta E_{\text{SN}}$ caused by the quantized flux of NIR probe photons15 (see Methods for details). Therefore, a RDN of $-0.04$ would correspond to a complete removal of the vacuum fluctuations in the space-time segment sampled in the DX.

The coherent field transients $E_{\text{MIR}}(t_D)$ generated by optical rectification of a NIR 12-fs pump with a pulse energy of 3.5 nJ are depicted in Fig. 2a. Two waveforms with precisely inverted amplitudes result from rotation of the pump polarization by 90° around the optical axis (black and grey lines). The broadband amplitude spectra (inset) have an average frequency of 44 THz, corresponding to a free-space wavelength of 6.8 μm and photon energy of 180 meV. Figure 2b shows the RDN amplitudes recorded simultaneously. Dark (light) blue areas denote delay times with negative values induced by the black (grey) transient in Fig. 2a, indicating a clear squeezing of the local electric field fluctuations $\Delta E_{\text{rms}}$ below the level of the bare quantum vacuum. Time segments carrying excess noise with respect to the vacuum ground state are filled in red. Salient features in the noise patterns in Fig. 2b are evident. First, there is a clear asymmetry, with positive excess noise surpassing the absolute values of vacuum squeezing, especially in the region close to the centre of the transients where the amplitudes...
are greatest. Second, the noise maxima in Fig. 2b coincide with the maximally positive slopes of the coherent field amplitudes in Fig. 2a, while optimum squeezing of $\Delta E_{\text{rms}}(t_0)$ is obtained close to the positions with a maximal decrease in $E_{\text{MIR}}(t_0)$ with time (see vertical dashed lines). Finally, owing to this inherent polar asymmetry in $\Delta E_{\text{rms}}$, the shift in carrier-envelope phase between the black and grey transients in Fig. 2a results in distinctly different quantum noise patterns that are not mirror images of each other.

We now investigate the physical origin of these findings. We first vary the pulse energy in the NIR pump, which is proportional to the electric field amplitude $E_{\text{MIR}}$ and record the resulting RDN amplitudes (Fig. 3). At low pump energies of 0.8 nJ and 1.5 nJ, the noise patterns are still fairly symmetric with respect to positive and negative extrema. The asymmetry towards positive excess noise appears clearly at 2.5 nJ (Fig. 3). At low pump energies of 0.8 nJ and 1.5 nJ, the noise patterns are still fairly symmetric with respect to positive and negative extrema. The origin of these observations is qualitatively understood in terms of the following expression for $\Delta E_{\text{rms}}(t)$ at the exit surface of a GX (see Methods for details):

$$\Delta E_{\text{rms}}(t) = e^{f(t)} \Delta E_{\text{vac}}$$  

(1)

where

$$f(t) = \frac{dt}{nc} \frac{\partial E_{\text{MIR}}(t)}{\partial t}$$  

(2)

denotes the squeezing factor in the time domain. We adopt plane waves and negligible pump depletion in a medium with a second-order nonlinear coefficient $d$ and thickness $l$. The bare vacuum amplitude $\Delta E_{\text{vac}}$ is assumed as input, which is adequate for the quantum properties of the coherent pump. $c$ denotes the speed of light in vacuum and the assumption of a constant refractive index $n$ is well justified because of the minor dispersion of GaSe in the MIR. It is evident from equations (1) and (2) that the extrema in $\Delta E_{\text{rms}}(t)$ are expected at the positions of maximum slope of $E_{\text{MIR}}(t)$, as confirmed experimentally in Fig. 2. With increasing $E_{\text{MIR}}(t)$, a nonlinear relationship between squeezing and excess noise, referenced to $\Delta E_{\text{vac}}$, results because of the exponential character of equation (1). This relationship tentatively explains the build-up of the asymmetry in Fig. 3. We now select two points in time, $t_{\text{max}}$ and $t_{\text{min}}$, at which the slope of $E_{\text{MIR}}(t)$ differs only by sign: $f(t_{\text{max}}) = |f(t_{\text{max}})| = f(t_{\text{min}})$. With equations (1) and (2) and the quantitative expression for the vacuum amplitude $E_{\text{vac}}$, we obtain

$$\Delta E_{\text{rms}}(t_{\text{max}}) \Delta E_{\text{rms}}(t_{\text{min}}) = (\Delta E_{\text{vac}})^2 = \frac{h}{\varepsilon_0 \Delta x \Delta y \Delta z \Delta t}$$  

(3)

where a four-dimensional space-time segment is defined by the transverse modal cross-section $\Delta x \Delta y = w_{\text{probe}}^2$ and the effective spatio-temporal length $\Delta z \Delta t$ that is set by the intensity envelope of the probe, $h$ is the reduced Planck constant and $\varepsilon_0$ is the permittivity of free space. To experimentally verify equation (3), two delay times with extremal time derivatives of $E_{\text{MIR}}(t)$ are sampled. We plot the measured values for RDN($t_{\text{max}}$) and RDN($t_{\text{min}}$) versus NIR pump pulse energy (red and blue circles in Fig. 4, respectively). The green curves represent a least-squares fit to the data based solely on equation (3). Saturation behaviour is found for squeezing and a superlinear increase is found for anti-squeezing, demonstrating good agreement between experiment and theory.
agreement with the fact that there is only one unique ground state. This is in the symmetric deviations from the vacuum level detected under low noise patterns therefore correspond to an ultrabroadband generation of analogous to established squeezing experiments based on a second-order squeezing inside the GX inferred by the analysis and a resulting RDN non-classical state with bare vacuum noise. Therefore, the 50% of local matching to the spatio-temporal probe wave packet contaminate the quantum field that is achieved inside the GX. Spurious reflections at the uncoated surfaces of the GX and DX as well as imperfect segment thickness of 0.04 that would result under complete suppression of MIR vacuum signals. Traditionally, squeezing has been considered in the frequency domain, and is attributed to amplification and de-amplification of specific field quadratures. But the electro-optic effect motivated in the introduction links the $\partial E_{\text{MIR}}/\partial t$ term in equation (2) to a modulation of the refractive index $\partial n/\partial t$. Therefore, redistribution of vacuum fluctuations following local advancement and slowdown of the speed of light represents an attractive alternative with which to illustrate generation of non-classical radiation in our subcycle time-resolved situation. The time-domain manifestation of Heisenberg’s uncertainty principle in equation (3) leads to an imbalance between the excess noise related to acceleration of the co-propagating reference frame and squeezing of the quantum amplitude that originates from local deceleration (see Methods).

In conclusion, a time-domain perspective on quantum electrodynamics works with subcycle resolution and direct referencing of electric field fluctuations to the quantum vacuum. The high peak intensities provided by few-femtosecond laser pulses of minute energy content enable a compact quantum technology based on broadband nonlinearities without immediate need for enhancement cavities, waveguides or cryogenic cooling. Many fundamental questions arise concerning a generalized understanding of quadratures being linked to local accelerations of the moving reference frame and regarding the benefits and limitations of the inherently non-destructive character of the technique. Future extensions could aim to achieve, for example, a full quantum tomography on subcycle scales. Filling the gap created by a lack of quantum approaches in the MIR or multi-terahertz range provides interesting perspectives: access to new quantum states produced by subcycle perturbation of ultrasonically coupled light matter systems might be one application; quantum spectroscopy and manipulation of collective degrees of freedom in condensed matter are also inherent to this regime.

Online Content Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.
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Author Contributions A.L. conceived the idea and wrote the first version of the paper. C.R., D.V.S. and A.L. designed the experiment. C.R., P.S., M.S. and D.V.S. performed the measurements and analysed the data. A.S.M. and Q.B. worked out the theoretical description. All authors discussed the results and contributed to the writing of the final manuscript.

Author Information Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to A.L. (aleitens@uni-konstanz.de).

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THEORETICAL CONSIDERATIONS LEADING TO EQUATIONS (1) AND (2). The generation of the quantum electric field patterns in our experiment can be understood as a series of two subsequent nonlinear processes of second order. First, a few-femtosecond pump pulse in the NIR produces an ultrashort and coherent electric field transient $E_{\text{MIR}}(t)$ at MIR frequencies $\Omega$ by optical rectification in a GX. This step corresponds to a difference-frequency mixing process within the broadband spectrum of the pump, resulting in an identical carrier-envelope envelope phase for MIR transients in the entire pulse train produced by the mode-locked laser system.\(^{10}\)

In the second step, $E_{\text{MIR}}(t)$ starts driving the second-order nonlinearity in the GX. We adopt propagation of plane waves in the nonlinear element along the $z$ axis from $-l/2$ to $l/2$ and an appropriate mutual orientation of pump field polarization and GX. The one-dimensional picture is well justified in our geometry because the thicknesses of both the GX and DX are smaller than the Rayleigh length of the MIR radiation that is generated. Together with the high-NA off-axis parabolic mirrors, this fact ensures proper matching to a single transverse mode. The nonlinear coefficient $d$ is proportional to the second-order nonlinearity $\chi^{(2)}$ of the emitter material;\(^{10}\) it is the linear refractive index. All susceptibilities may be assumed to be dispersionless when the MIR frequencies $\Omega$ are far from the electronic and optical phonon resonances of the medium. In the vacuum picture,\(^{33}\) the total MIR quantum field can be written $E_{\text{MIR}} = E_{\text{H}} + E_{\text{EMIR}}$, with the coherent amplitude $E_{\text{H}} = \langle E_{\text{H}} \rangle$ and a pure quantum correction $E_{\text{EMIR}}$. Locally, $E_{\text{EMIR}}$ induces the second-order nonlinear polarization $P_{\text{EMIR}} = -dE_{\text{EMIR}}$, which acts as a source in the wave equation. We restrict ourselves to small pump depletion by omitting the correction to the coherent part and neglect the second-order terms in $E_{\text{EMIR}}$. The slowly varying amplitude approximation\(^{33}\) then leads to

$$\frac{\partial E_{\text{EMIR}}(z, \Omega)}{\partial z} = -\frac{\Omega d}{nc} \int E_{\text{MIR}}(z, \Omega') - E_{\text{EMIR}}(z, \Omega') d\Omega'$$

Transforming back into the time domain and using a retarded reference frame with $t = t - c(z - \Omega z)$, i.e., $E_{\text{EMIR}}(z', t') = \delta E_{\text{EMIR}}(z', t')$ and $E_{\text{MIR}}(z', t') = E_{\text{MIR}}(z, t)$, we obtain

$$\frac{\partial \delta E_{\text{EMIR}}}{\partial t'} = \frac{d}{nc} \int \frac{\partial E_{\text{MIR}}}{\partial t'} \delta E_{\text{EMIR}}(z', t') + E_{\text{MIR}}(z', t') \frac{\partial \delta E_{\text{EMIR}}}{\partial t'} dt'$$

As long as deviations of the quantum field from the level of bare vacuum remain moderate, the temporal derivative of $\delta E_{\text{EMIR}}$ is negligible and we can omit the second term in the braces on the right-hand side of equation (5). The same term vanishes even for large squeezing when $E_{\text{MIR}}(z', t')$ is sufficiently small. In both cases, an analytical solution of the partial differential equation is straightforward by integrating over $z'$. Returning to the original reference frame, the field at the exit surface of the GX, $E_{\text{MIR, out}}(t) \equiv E_{\text{MIR, out}}(z = l/2, t)$, can be expressed as $E_{\text{MIR, out}}(t) = e^{i\Omega t} \delta E_{\text{MIR}}(l/2, t)$, where $\delta E_{\text{MIR, out}}(t) \equiv \delta E_{\text{MIR, out}}(z = l/2, t)$ and

$$f(t) = \frac{dE_{\text{MIR}}(z = l/2, t)}{dt}$$

reverses equation (2). Then, calculating the r.m.s. standard deviation $\Delta E_{\text{MIR}}(t) \equiv \langle \delta E_{\text{MIR, out}}(l/2, t)^2 \rangle^{1/2}$ at the end of the nonlinear section results in

$$\Delta E_{\text{MIR}}(t) = f(t) \left( \frac{\delta E_{\text{MIR, in}}^2}{t} \right)^{1/2}$$

Equation (1) follows from a bare vacuum or fully coherent input, as in our experiment. In this case, $\langle \delta E_{\text{MIR, in}}^2(l/2, t) \rangle$ is given by the r.m.s. vacuum electric field $E_{\text{vac}}$. Time-domain noise patterns and temporal changes of the local phase velocity. The Pockels effect\(^{15}\) causes a change in refractive index $\Delta n = n' E_{\text{MIR}}$, with the effective electro-optic coefficient $r = -n' dE_{\text{MIR}}/dt$ and therefore $f(t)$ to accelerations and retardations of the local reference frame. The linear refractive index is defined as the ratio between the velocity of light in vacuum $c$ and the local phase velocity $\nu_{\text{loc}}$: $n(t) = \nu_{\text{loc}}(t)$. Together with equation (2), we find

$$f(t) = \frac{1}{c} \frac{\partial n}{\partial t} = \frac{n^2}{c^3} \frac{\partial \nu_{\text{loc}}}{\partial t}$$

(6)

to first order in $\Delta n$. This expression is of general character because it does not depend on the specific nonlinearity used to induce the phase shifts that result in squeezing of the electromagnetic field and ultimately the emission of non-classical radiation. For example, analogous noise patterns to those found in our experiments might result from direct modulation of $\nu_{\text{loc}}$ by the NIR pump intensity $I_p(t)$ via...
third-order effects causing a nonlinear index of refraction $n_2$ and therefore $\Delta n(t) = n_2 J(t)$. It is clear from equations (1), (2) and (6) that excess noise with respect to the bare vacuum level is linked to acceleration of the local reference frame, that is, $\partial v_{loc}/\partial t > 0$. On the other hand, retardation with $\partial v_{loc}/\partial t < 0$ underlies a decrease in the local quantum fluctuations. These facts lead us to suggest a generalized understanding of quadratures in a time-domain context, as outlined in the conclusion of the main text.

**Data availability.** The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Extended Data Figure 1 | Sketch of the complete set-up underlying the experiments. A mode-locked Er:fibre laser oscillator (Er:OSC) forms a train of femtosecond pulses at a repetition rate of 40 MHz (green). After a fibre-optic beam splitter, an electro-optic modulator (EOM) removes every second pulse in the pump branch (top), reducing the repetition rate to 20 MHz, and 12-fs pulses of centre wavelength $\lambda_c = 1,550$ nm are generated by an Er:doped-fibre amplifier (pump EDFA) with subsequent pulse compression (orange pulse train). A pulse energy $E_{\text{pump}} = 3.5$ nJ is applied to pump the GaSe generation crystal (GX). The pulse train seeding the probe amplifier (probe EDFA) is left at a rate of 40 MHz and compressed to a duration of 5.8 fs and centre wavelength $\lambda_c = 1,150$ nm (blue pulse train). Eventually, a pulse energy of 120 pJ from this branch is available for probing the electro-optic effect in the AgGaS$_2$ detector crystal (DX). Details on the laser technology are found in ref. 30. The NIR pump beam is focused into the GX by an off-axis parabolic mirror (sketched in yellow). A MIR electric field transient (red beam path) is generated by the second-order nonlinearity of the GX, collimated by another parabolic mirror and sent through a thin GaSb wafer acting as low-pass filter (LPF). This component is inserted at Brewster’s angle for minimum loss of the MIR field and complete suppression of the NIR pump. The probe pulse train travels over a variable optical delay stage (VD) to set the relative timing $t_D$ between pump and probe. It is reflected as s-polarized at a silicon beam combiner (BC), which is also inserted at Brewster’s angle with respect to the p-polarized MIR field. Both beams are focused to co-propagate inside the DX. The probe pulse train is then collimated by a final off-axis parabola and sent through an ellipsometer that consists of an achromatic quarter-wave plate ($\lambda/4$) and a Wollaston prism (WP). A balanced pair of photodiodes (BPD) detects a differential photocurrent, which is proportional to the MIR electric field amplitude that has been co-propagating with the probe. In total, only $E_{\text{probe}} = 80$ pJ of probe pulse energy arrives at the BPD, owing to optical losses. Signals are demodulated at the Nyquist frequency of 20 MHz by a radio-frequency lock-in amplifier (RF lock-in) referenced to the EOM that picks every second pulse out of the pump pulse train. Signal collection and control of the VD are carried out by a personal computer (PC). An in-depth evaluation of the electro-optic sampling scheme is found in ref. 29.
Extended Data Figure 2 | Explaining the concept of differential quantum sampling. **a**, Lock-in demodulation scheme for obtaining coherent amplitude and RDN. Individual readout values for the differential photocurrent $\Delta I/I$ are sketched as a function of real time. The readouts carrying signal due to pumping of the GX are red and the sub-pulse train sampling bare vacuum input is shown in blue. The time interval of 25 ns between each probe pulse is given by the inverse of the probe repetition rate of 40 MHz. Demodulation of the coherent field amplitude $E_{\text{MIR}}$ is visualized in the top panel: the phase of the lock-in measurement at a reference frequency of 20 MHz is set such that the maximum difference between the average amplitude in the signal pulse train (red shading) and the reference pulse train (blue shading) is detected. Operating at the Nyquist frequency of the probe pulse train offers optimal performance in terms of amplitude and timing stability. Measurement of the RDN is illustrated in the bottom panel: we exploit two orthogonal readout phases at a reference frequency of 10 MHz to determine the level of fluctuations in the signal (red shading) and reference pulse train (blue shading) is detected. In both cases, the standard deviation is computed; it corresponds to a convolution of the identical shot noise $\Delta E_{\text{SN}}$ in each channel and the noise due to the quantum fluctuations of the MIR electric field, which includes the pump-induced modulation of the quantum noise $\Delta E_{\text{rms}}$ in the signal channel and the bare vacuum noise $\Delta E_{\text{vac}}$ in the reference channel. The sketch on the right-hand side corresponds to a delay time with strong anti-squeezing: here the r.m.s. width of the noise distribution in the signal channel (red) is larger than that of the reference noise (blue). This case would result in a positive value for the RDN. **b**, Individual readouts from the signal and reference channels. Raw data of a single scan over delay time $t_D$ from the two separate noise channels of the differential detection scheme, collected within 150 s. The blue curve results from the reference channel, which has a radio-frequency phase set such that only the standard deviation of electro-optic readouts $\Delta I/I$ with no pump pulse present in the generation crystal is probed at a reference frequency of 10 MHz (blue sub-pulse train and dashed line in **a**). Detecting only the bare vacuum input, this channel shows no dependence on the relative time delay $t_D$ between pumping and probing. In contrast, the channel with radio-frequency phase adjusted to sample the relative noise in the sub-pulse train that contains signals from the emitter excited by a pump pulse is plotted in red (noise signal, phase offset of 90° with respect to the reference channel; see red pulse train and line in **a**). Positions showing strong overshoot noise with respect to the vacuum reference are evident and indicate ‘anti-squeezing’, as sketched in an exaggerated way on the right-hand side of the bottom panel in **a**. But also delay times with the noise level of the red signal channel falling substantially below the blue reference data are clearly discernible, corresponding to ‘squeezing’ of the electric field with respect to the bare quantum vacuum. The data depicted in Figs 2 and 3 were processed in real time in the differential way described by equation (4) to obtain the RDN, subtracting any small variations that occur in the absolute noise floor of the laser or electro-optic detector. Statistics may be enhanced by averaging over multiple such scans.
Extended Data Figure 3 | Radio-frequency noise spectrum of the electro-optic detector. The relative noise spectral density at the output of the differential photodetector BPD, that is, at the input of the radio-frequency lock-in amplifier (see Extended Data Fig. 1), is shown. The black curve was recorded with the probe beam blocked and represents the electronic background (dark) noise of the system. The blue curve shows the noise spectrum under full illumination of the balanced photodiodes by the probe pulse train of repetition rate 40 MHz, but in the absence of any electro-optic signal input. The electronic low-pass filter is designed for a broad plateau of constant amplification and flat phase around the centre frequency of 10 MHz used for detection of the RDN (see Extended Data Fig. 2). The roll-off at a frequency of 20 MHz still leaves sufficient sensitivity for low-noise collection of the mean signal amplitudes, but results in a change in the radio-frequency phase angle with respect to the noise detection at 10 MHz, which needs to be adjusted correctly. The optical phase angle between the measured coherent field transients and the RDN patterns is not affected by this dispersion, because it results exclusively from scanning of the optical delay time \( t_D \). The red dashed line represents the relative shot-noise level calculated for a Poisson input of \( 5 \times 10^8 \) photoelectrons generated per probe pulse. It demonstrates that the broad noise plateau around 10 MHz is dominated by the shot noise of the probe photon flux, ensuring independence of the RDN signal from the measurement bandwidth set by the lock-in amplifier. Strong suppression of the 40 MHz signal from the fundamental pulse train is evident from the small maximum in the blue curve and helps to ensure adequate linearity of the electronic system.
Extended Data Figure 4 | Excluding nonlinearities in the readout electronics and technical noise of the coherent transients as potential spurious sources of RDN signals. A universal check for any influence in the noise readout due to the presence of finite coherent field amplitudes is as follows. We exchange the roles of the two photodiodes at the input of the differential detector by rotating the Wollaston prism (WP) in the ellipsometer by an angle of 180° (see Extended Data Fig. 1). In this way, the polarity of the classical electric transients is inverted electronically but not physically (in contrast to Fig. 2, in which the pump polarization on the generation crystal has been rotated by 90°), resulting in the blue and red curves in the top panel. If the differential noise traces depicted in the bottom panel were artefacts due to some response of the electronic noise detection to either the amplitude or the slope of the mean readout, then we would expect those signals to also invert sign, but this is not the case. The high degree of agreement between the blue and red noise traces in the bottom panel, corresponding to the original and electronically inverted transients in the top panel, is a good indication of the linearity of our readout scheme. The result of this procedure implies that our noise traces must have an optical origin and that they do not result from any spurious processes in the readout electronics. The noise patterns in the bottom panel also cannot result from amplitude or phase noise of the coherent transients: in both cases, the periodicity of the noise would follow the second harmonic of the coherent transients and no negative signals with respect to bare vacuum noise would be expected.