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Ultrafast all-optical switching by single photons

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An as yet outstanding goal in quantum optics is the realization of fast optical nonlinearities at the single-photon level. This would allow for the implementation of optical devices with new functionalities such as single-photon switches/transistors[1,2] or controlled-phase gates[3]. Although nonlinear optics effects at the single-emitter level have been demonstrated in a number of systems[4–13], none of these experiments showed single-photon switching on ultrafast timescales. Here, we perform pulsed two-colour spectroscopy and demonstrate that, in a strongly coupled quantum dot–cavity system, the presence of a single photon on one of the fundamental polariton transitions can turn on light scattering on a transition from the first to the second Jaynes–Cummings manifold. The overall switching time of this single-photon all-optical switch[14] is ∼50 ps. In addition, we use the single-photon nonlinearity to implement a pulse correlator. Our quantum dot–cavity system could form the building block of future high-bandwidth photonic networks operating in the quantum regime[15–18].

A single-photon switch is a device in which a single optical gate (or control) photon controls the propagation (or scattering) of incident signal photons by means of nonlinear optical interactions[1] (Fig. 1a). The requisite large photon–photon interactions are provided by the basic system of cavity quantum electrodynamics (QED)[19], namely that of a single quantum emitter strongly coupled to a single cavity mode. Figure 1b presents the first two rungs of the corresponding energy diagram, the so-called Jaynes–Cummings ladder. The anharmonicity of the Jaynes–Cummings ladder and the resulting single-photon nonlinearity is a result of the quantized nature of the radiation field and has been demonstrated either spectroscopically or through photon-correlation measurements for almost all cavity-QED implementations, including atoms coupled to Fabry–Pérot or toroidal microcavities[4–6,20], super-conducting qubits in strip-line resonators[7–9,21] or quantum dots (QDs) in nanocavities[11–14,22]. However, the speed of devices based on single-photon nonlinearities has so far not been addressed. As the ultimate switching times are limited by the reciprocal emitter–cavity coupling strength, QDs strongly coupled to nanocavities[10,23–25] have emerged as ideal candidates for the realization of ultrafast single-photon nonlinear devices because of their record-high coupling strengths.

Our device consists of an InAs/GaAs QD positioned at the electric-field maximum of a photonic-crystal defect cavity in L3 geometry[10]. The QD–cavity system is deep in the strong coupling regime. With a coherent coupling constant g of 141 μeV and a quality factor of $Q \approx 25{,}000$ (that is, a cavity photon decay rate κ corresponding to 53 μeV), the figure-of-merit for the anharmonicity of the coupled system, $g/\kappa \approx 2.7$, well exceeds the previously reported values in the literature. Resonant spectroscopy of the strongly coupled system was performed using a crossed-polarization technique, which ensures efficient suppression of the excitation-laser light back-reflected from the sample surface[22]. Ultrafast laser pulses with pulse durations between 33 and 86 ps were derived from a mode-locked Ti:sapphire laser (see Methods). The pulse delay between control and signal pulse was adjusted using a motorized delay stage (Fig. 1c).

We first carried out resonant spectroscopy of the strongly coupled QD–cavity system by scanning the central frequency of the pulsed laser across the polariton spectrum (see Methods) when QD and cavity were very close to resonance. The result of a single scan is displayed as open circles in Fig. 2a for a pulse duration of 86 ps and an average signal power of 1 nW. As reported previously[22], we used an off-resonant re-pump laser at a repetition rate of 1 MHz to partially counteract the laser-induced QD blinking present in the system. As a result of this blinking, we obtained a three-peak spectrum consisting of upper and lower polariton peaks (UP and LP) as well as the uncoupled cavity peak in the middle (see section ‘Estimating typical blinking times’ in the Supplementary Information). To ensure that the switching operation of our device was enabled by the presence of a single photon in the cavity, we carried out photon autocorrelation measurements[22] demonstrating photon antibunching (that is, photon blockade[4]) on both upper and lower polariton transitions to the first Jaynes–Cummings manifold (see section ‘Photon correlation measurements’ in the Supplementary Information).

We next performed pulsed two-colour spectroscopy of the strongly coupled QD–cavity system. The control laser was tuned (average power of 2 nW) to the upper polariton resonance, and the signal laser pulse was scanned across the spectrum. Note that the power of the control laser corresponds to ∼120 photons per pulse. With a laser–cavity coupling efficiency of ∼2% and a 50% photon content for the polaritons, this yields, on average, ∼1.2 photons per pulse scattered off the strongly coupled system. For the scan, the delay of the signal pulse with respect to the control pulse was chosen to be 25 ps, corresponding to the polariton lifetime. The resulting spectrum is also displayed in Fig. 2a, with the filled circles showing the system response with control laser present, and the open circles the response without control laser. The difference between the two data sets directly reflects the nonlinear response of the QD–cavity system, with the reduction of the polariton signal being the most obvious effect of the presence of the control laser. This difference is plotted in Fig. 2b (red filled circles). In addition to the fast photon–photon interactions of interest, the coupled system also exhibits a slow nonlinearity caused by the laser-induced QD blinking discussed in the previous paragraph (see section ‘Estimating typical blinking times’ in the Supplementary Information). To distinguish between the two effects, we acquired the same spectra as in Fig. 2a, but now with a time delay of 5 ns between the control and signal pulse, which is much longer than the polariton lifetime and the laser pulse

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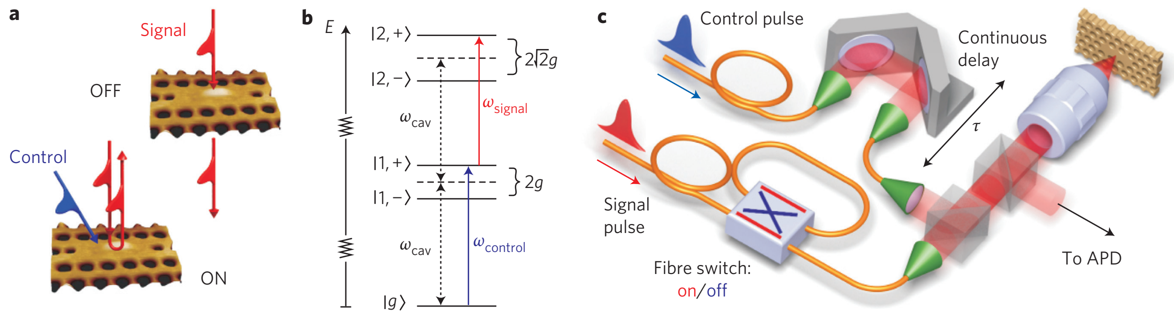

Figure 1 | A single-photon all-optical switch. a, A single control photon incident on the QD-cavity device determines whether a signal photon of different colour is scattered. **b**, Energy-level diagram of the strongly coupled QD-cavity system up to the second manifold of the anharmonic Jaynes–Cummings ladder. A single control photon on the upper polariton transition (UP) to the first manifold ($|g\rangle \rightarrow |1,+\rangle$) changes the scattering rate of a second signal photon resonant with a transition from the first to the second manifold ($|1,+\rangle \rightarrow |2,+\rangle$). **c**, Set-up for the demonstration of ultrafast single-photon switch operation. The relative delay between signal and control pulses is adjusted by a continuous delay stage. In addition, a discrete delay line, corresponding to a time delay of ∼5 ns, can be added to the path of the signal pulse using a fibre switch. The photons back-scattered from the QD-cavity system are detected by an avalanche photodiode (APD) in single-photon counting mode.

durations. The difference signal is displayed in Fig. 2b as blue filled circles. Subtracting the two nonlinear responses in Fig. 2b from one another yields the fast nonlinear optical response from the strongly coupled QD–cavity system, as presented in Fig. 2c. The data show that the largest nonlinear effect occurs at the spectral position of the polaritons (vertical blue lines). Here, the change in the scattering rate induced by the control laser is negative (∼–15%) due to saturation of the corresponding transitions. At the transition wavelength from the first to the second manifold (vertical red line), this change is positive (∼+6%), because the absorption of the control photon enables the subsequent scattering of a signal photon. The relative increase in reflection signal is rather moderate due to the presence of the uncoupled cavity peak. In the absence of blinking and background laser light, we theoretically expect a switching contrast of +110%. In addition to the contribution from the transition of interest, we also observe a non-negligible positive response at slightly positive detunings, which we attribute to pure exciton dephasing (see section 'Pure exciton dephasing' in the Supplementary Information). To compare our experimental data with theoretical expectations, we performed numerical simulations based on a Monte Carlo wavefunction (MCWF) approach, with the experimental parameters as input and only the absolute amplitude of the nonlinear signal determined from a least-squares fit to the data (see section 'Calculation of the optical non-linearity' in the Supplementary Information). The excellent agreement between theory (black line) and experiment clearly demonstrates that the observed positive nonlinearity is indeed due to the two-colour transition to the second Jaynes–Cummings manifold, ensuring the single-photon nature of the observed nonlinearities. We emphasize here that, because of the finite linewidth of the coupled-system eigenstates as well as the finite bandwidth of the laser pulses, there is some overlap between transitions from the first to the second and from the second to the third manifold, and so forth. Hence, we expect a non-negligible contribution to the nonlinear signal stemming from states higher up in the Jaynes–Cummings ladder.

To demonstrate ultrafast switching by single photons—that is, conditional scattering of signal photons on ultrafast timescales—we varied the delay between control and signal pulses while recording the (positive) nonlinearity for fixed laser-detunings. As depicted in Fig. 1b, the control pulse was chosen to be resonant with the fundamental upper polariton transition and the signal pulse to be

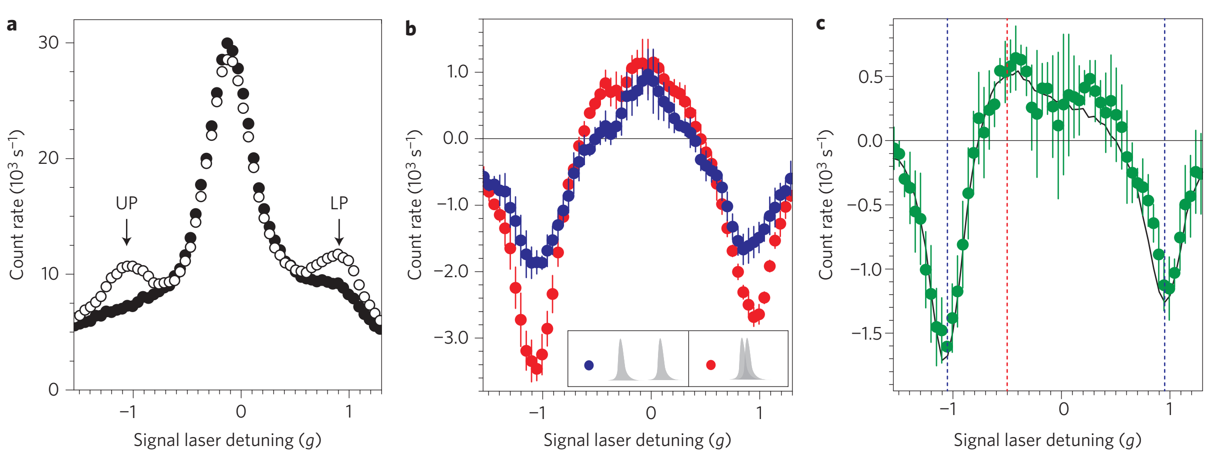

Figure 2 | Two-colour spectroscopy of the strongly coupled QD-cavity device. a, System response with (filled circles) and without (open circles) control laser present when scanning the signal laser across the QD-cavity spectrum (for a cavity detuning of ∼−0.1g). Pulse durations of both control and signal laser pulses were 86 ps. **b**, Nonlinear behaviour is observed for a time delay of 5 ns (blue filled circles) and 25 ps (red filled circles) between the control and signal pulses. **c**, Subtracting the red and blue data points of **b** from one another, we obtain the system nonlinearity due to the Jaynes–Cummings dynamics, which is taking place on ultrashort timescales. The red vertical line indicates the transition from the first to the second manifold ($|1,+\rangle \rightarrow |2,+\rangle$). The positions of the fundamental polaritons are marked by blue vertical lines. In **b** and **c**, three adjacent data points were averaged. The black curve in **c** was obtained from a MCWF simulation of the system dynamics with the experimental parameters as input.

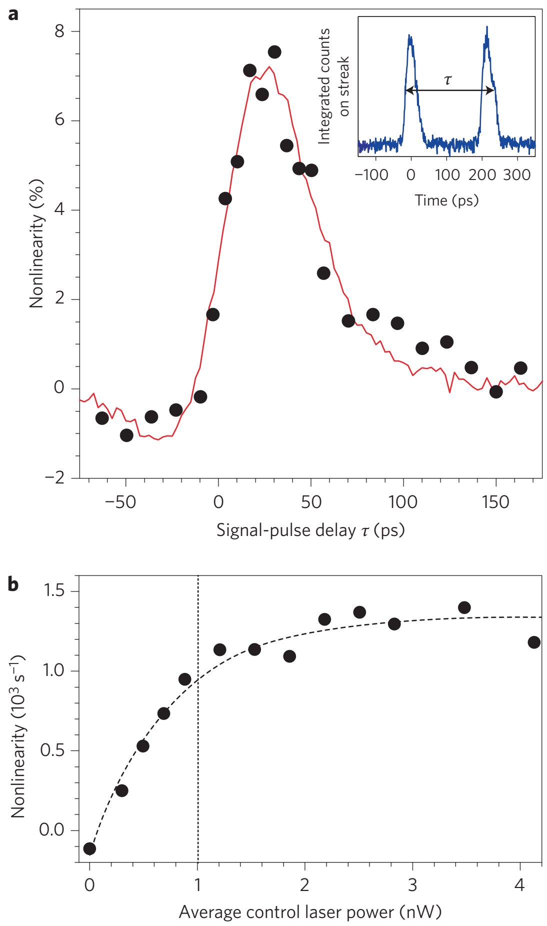

Figure 3 | Ultrafast nonlinear response. a, Measured nonlinear response of the QD–cavity system (black filled circles) as a function of delay between control and signal laser pulses for a pulse FWHM of 33 ps. Here, the nonlinearity is given as the relative increase/decrease of the detected photon scattering rate compared to the case without control laser. The red curve was obtained from a numerical simulation using a MCWF approach with the experimental parameters as input. We observe a slight systematic deviation of the data from the theoretical expectation for time delays between 60 ps and 150 ps. One possible explanation is the indirect excitation of an additional (slow) excitonic mode that is very weakly coupled to the cavity mode due to the slight mismatch between the *x*–*y* polarization axis of the QD and the cavity axis. **b**, Transfer characteristic of the single-photon switch. The nonlinear signal was recorded as a function of control power for a pulse delay of 25 ps. For average powers larger than 1 nW, the system saturates. The dashed line is a guide to the eye.

resonant with the transition from the first to the second manifold. The result is plotted in Fig. 3a for a pulse duration of 33 ps; the black filled circles represent experimental data, and the red curve was obtained from a MCWF simulation with the absolute amplitude extracted from a least-squares fit to the data. Note that the nonlinearity is plotted directly as a relative increase/decrease of the detected signal photon scattering rate, compared to the case without control laser corresponding to the figure-of-merit of our switch. As the data show, our single-photon switch combines both ultrafast turn-on and turn-off times, a combination that is typically hard to achieve in other quantum emitter–cavity systems. The sharp turn-on of the nonlinear system response around zero time delay is directly related to the parameters of the laser pulses. We find that in this particular case the corresponding turn-on time (the time the signal takes to rise from 10% to 90% of the maximum) is ~20 ps. This is very close to the ultimate switch-on time, which is limited by the anharmonicity of the Jaynes–Cummings spectrum, and is given by $((2-\sqrt{2})g)^{-1} \approx 20$ ps. For shorter pulse durations the spectral selectivity of the control laser and therefore the performance of the switch would be significantly degraded.

At delay times longer than ~30 ps, the nonlinear signal of Fig. 3a exhibits a fast decay with a $1/e$ time of ~30 ps, close to the polariton lifetime of 25 ps, which in turn sets the fundamental limit for the turn-off time of our device. The asymmetry in the pulse-delay dependence is a direct consequence of the cascaded nature of the underlying two-photon transition (Fig. 1b) and is most pronounced for pulse durations on the order of the polariton lifetime.

As well as the switching speed, a quantity of interest is the transfer characteristic, that is, the output signal as a function of control power, which is plotted in Fig. 3b. Here, we recorded the number of signal photons scattered from the system as a function of input power of the control pulse. As expected, we observe an initial linear increase with control laser power, and then saturation of the polariton transition corresponding to a mean intracavity average photon (polariton) number of ~0.25 (0.5).

Besides the realization of single-photon switching, the strong nonlinearity of the QD–cavity system can be applied to measuring

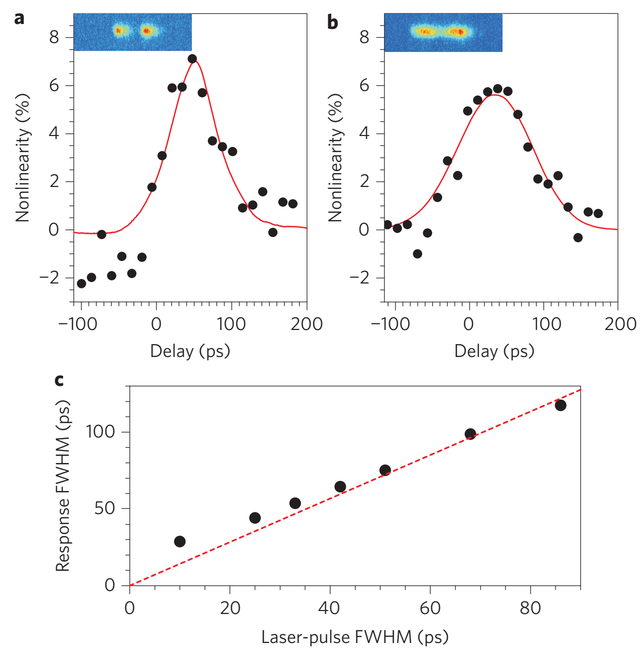

Figure 4 | Pulse correlator. a,b, The strength of the nonlinearity, as defined in the caption of Fig. 3, as a function of delay for pulse durations of 51 ps (**a**) and 86 ps (**b**). The longer the pulse, the more symmetric the nonlinear response. The red curves are the numerical convolutions of the streak-camera images shown as insets. The absolute amplitudes and the peak positions of the convoluted signals were fit to the data. In both cases, control and signal laser had an average power of 1 nW. This power corresponds to ~60 photons per pulse impinging on the sample surface or a pulse energy of 80 eV, which means that on average 0.6 photons per pulse are scattered off the system. **c**, Simulated width (filled circles) of the nonlinear system response versus laser pulse width. For pulse durations larger than 50 ps, the simulated width of the nonlinear signal approaches that of the correlator width of the laser pulses (red dashed line). For very short pulses, the polariton lifetime sets the lower limit for the system response.

pulse durations of ultrafast optical pulses down to the single-photon level. Pulse-correlator operation is demonstrated in Fig. 4, where the nonlinear system response is mapped as a function of pulse delay for pulse durations of 51 ps and 86 ps. Because the pulse durations are significantly longer than the polariton lifetime of 25 ps, the system response is more symmetric than was the case for the 33 ps pulses of Fig. 3a. We find good agreement between the pulse-delay-dependent nonlinearity and the numerical convolutions of the independently obtained streak camera images (red lines).

Figure 4c compares the full-width at half-maximum (FWHM) obtained from Monte Carlo simulations of the nonlinear system response with the FWHM of the incident (Gaussian) laser pulses. The red dashed line corresponds to the correlator width of the pulses. Above 50 ps, the deviation of the simulated width from this line is less than 5%, so in this range our device works nicely as a pulse correlator. Finally, we remark that single-photon switch and/or pulse-correlator operation can also be realized by centring the signal pulse on the other (lower) polariton transition, which yields a larger magnitude for the nonlinearity (Fig. 2c; see section 'Cross-correlations of upper and lower polaritons' in the Supplementary Information).

A natural extension of our work would be the realization of a single-photon transistor, in which the presence of a single control photon ($N_c = 1$) enables the scattering of $N_s \geq 2$ signal photons[1,2]. A simple calculation shows that if pure dephasing was absent (see section 'Transistor operation' in the Supplementary Information), our QD-cavity device would exhibit a modest gain of $G = N_s/N_c > 2$. While increasing the ratio g/κ will already increase G, high-gain ($G \gg 1$) transistors may be realized in combination with electromagnetically induced transparency (EIT) schemes[26–29]. Finally, combining our device with state-of-the-art waveguide technology[16–18] and implementing QD charge control using p–i–n structures[30] to suppress blinking would enable high-contrast all-optical switching of single-photon pulses. This might enable the demonstration of the preservation of quantum coherence during the nonlinear interaction, which in turn could pave the way for the realization of an ultrafast controlled-phase gate between two single-photon pulses[3].

After submission of this work, we became aware of two recent papers reporting on related experiments[31,32].

Methods

Pulse preparation. Both control and signal laser pulses were derived from the same mode-locked Ti:sapphire laser with a pulse repetition rate of 76.3 MHz and an intrinsic pulse width of a few picoseconds. The laser pulses were sent through a grating spectrometer for frequency filtering and split by a 50/50 beamsplitter. Both beams were then coupled into single-mode optical fibres. The resulting spectral width of the pulses could be adjusted from 0.04 nm to 0.015 nm by an additional slit in front of the spectrometer that determines the effective numerical aperture of the spectrometer. The pulses were nearly Fourier-limited, so we could adjust the pulse duration from ~33 ps to 86 ps. The central frequency of the signal pulse was mechanically tuned using a piezo-driven mirror holder in front of the fibre coupler, enabling coupling of different parts of the spectrum into the fibre. The central frequency of the pulse was monitored using a wavemeter, and a computer-controlled feedback loop allowed tuning. The average power of both control and signal laser beams was stabilized using acousto-optical modulators. The relative delay of the two pulses was adjusted using a motorized delay stage. Pulse shapes and delays were monitored by sending the light reflected from the sample surface to a streak camera with a time resolution of ~4 ps.

Extraction of the optical nonlinearity. When applying both a control pulse at time t and a signal pulse at time $t + \tau$ to the system, the time-integrated response can be written as $N_{\mathrm{both\ on}}(\tau) = N_{\mathrm{control}} + N_{\mathrm{signal}} + N_{\mathrm{nl}}(\tau)$, where N_{control} and N_{signal} denote the number of scattered photons when only a control or signal laser, respectively, are applied. $N_{\mathrm{nl}}(\tau)$ is the total optical nonlinearity, quantified as the number of additional scattered photons. Its origin is twofold—a fast (approximately picosecond) contribution arising from the anharmonicity of the Jaynes–Cummings ladder (JC) and a slow (approximately microsecond) contribution stemming from charge blinking—so $N_{\mathrm{nl}}(\tau) = N_{\mathrm{nl,JC}}(\tau) + N_{\mathrm{nl,blinking}}(\tau)$. If we choose an intermediate timescale τ_{int} on the order of nanoseconds, such that ps $\ll \tau_{\mathrm{int}} \ll$ µs, then $N_{\mathrm{nl,JC}}(\tau_{\mathrm{int}}) \approx 0$. For $\tau \ll$ µs, $N_{\mathrm{nl,blinking}}(\tau_{\mathrm{int}}) \approx N_{\mathrm{nl,blinking}}(\tau)$ and

$$N_{\mathrm{nl,JC}}(\tau) \approx N_{\mathrm{nl}}(\tau) - N_{\mathrm{nl}}(\tau_{\mathrm{int}})$$

where

$$N_{\mathrm{nl}}(\tau) = N_{\mathrm{both\ on}}(\tau) - N_{\mathrm{control}} - N_{\mathrm{signal}}$$

To determine $N_{\mathrm{nl}}(\tau_{\mathrm{int}})$, we chose $\tau_{\mathrm{int}} \approx 5$ ns by switching an additional delay line into the path of the signal laser. To eliminate long-time drifts we simultaneously measured $N_{\mathrm{both\ on}}(\tau)$, N_{control} and N_{signal} by switching the control and signal lasers on and off at 5 kHz and 10 kHz, respectively, and sorting the output photons accordingly.

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Author contributions
T.V. and A.R. conducted the experiments, analysed the data and performed the simulations. M.W. made essential contributions to the experiment in its early stages. A.B., K.J.H. and E.L.H. fabricated the structure that ensures maximal dot cavity coupling. T.V., A.R. and A.I. conceived the experiment, discussed the results and wrote the manuscript.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permission information is available online at http://www.nature.com/reprints. Correspondence and requests for materials should be addressed to T.V. and A.I.

Competing financial interests
The authors declare no competing financial interests.