FD3
17:00 – 17:15

# Quantum Control of Phosphorus Donor Rydberg States in Silicon

Stephen A. Lynch*, P. Thornton Greenland*, Alexander F. G. van der Meer†, Benedict N. Murdin‡, Carl R. Pidgeon§, Britta Redlich†, Nguyen Q. Vinh¶, and Gabriel Aeppli*
*London Centre for Nanotechnology, University College London, WC1H 0AH, United Kingdom.
Email: stephen.lynch@ucl.ac.uk
†FOM Institute for Plasma Physics Rijnhuizen, P.O. Box 1207, NL-3430 BE, Nieuwegein, The Netherlands.
‡Advanced Technology Institute, University of Surrey, Guildford GU2 7XH, United Kingdom.
§Heriot-Watt University, Department of Physics, Riccarton, Edinburgh, EH14 4AS, United Kingdom.
¶UC Santa Barbara, Institute for Terahertz Science and Technology, Santa Barbara CA 93106-4170, USA.

***Abstract*—We demonstrate the first observation of a THz photon echo. We exploit the photon echo as an experimental tool to investigate the quantum coherence properties of excited donor Rydberg states of phosphorus in silicon.**

## I. Introduction

In the last decade, there has been resurging interest in the physics of group-V donors in silicon. This has recently lead to a dramatic increase in activity in the research field. Much of this renewed interest stemmed from a proposal by Kane that silicon doped with group-V donors might be exploited to realise a quantum computer [1]. A related scheme involving group-V donors in silicon was also proposed by Stoneham [2]. Many experiments have been recently performed to investigate the feasibility of this scheme. These include measurements of both the spin lifetimes [3] and lifetimes of the donor Rydberg states [4]. The second important development that rekindled interest in this research field was the invention of the optically pumped silicon laser [5]. This breakthrough was the first demonstration of lasing in a silicon-based material, and to-date lasing at THz frequencies has now been demonstrated for all four Group-V donors [6]–[8]. Recent work also suggests the possibility of electrically pumped lasing in this material system [9]–[11].

In this paper, we will present convincing evidence for the first observation of a THz photon echo. The photon echo is a purely quantum mechanical effect and it has no classical interpretation. It is the optical analogue of the spin echo commonly observed in Nuclear Magnetic Resonance (NMR) or Electron Spin Resonance (ESR) experiments. Since the photon echo is a coherent quantum effect we needed a coherent source of THz radiation. The experiments described were performed using the FELIX free electron laser at the FOM institute at Nieuwegein in the Netherlands. A full description of the experiment can be found in our recently published paper [12]. The main cogent points, however, are summarized in this conference abstract.

## II. Theory

For typical donors in Si, the lowest energy Lyman series line is in the THz regime. In the case of phosphorus (P) the $1s(A_1)$ to $2p_0$ transition is at 34.2 meV, equivalent to 36.2 $\mu$m and 8.29 THz [13]. There are also smaller level splittings associated with the broken rotational symmetry in the solid. The orbitals have an order of magnitude larger spatial extent than those for hydrogen in vacuum: the $2p_0$ level, for example has an extent of around 10 nm, enclosing about 104 Si atoms, and is thus comparable in size to transistors already in commercial use. Previous frequency [14] and time-domain studies [4] have established the astonishing longevity of the excited states, with a population lifetime $T_1$ of 200 ps for the $2p_0$ state (due to 1 phonon emission augmented by intervalley and umklapp processes and corresponding oscillator quality factors of 2000 or more.

A two-level atom resonantly illuminated by the high intensity coherent light from a laser undergoes Rabi oscillations at a frequency given by $\Omega = F_0\mu_{12}/\hbar$ where $F_0$ is the electric field envelope of the light beam and $\mu_{12}$ is the transition dipole matrix element. For a pulse of finite duration, the excited state polarization that remains in the system after the pulse has passed varies sinusoidally with the pulse area, $A_P = \mu_{12}/\hbar \int F(t)dt$. If the laser is at resonance with the $1s(A_1)$ to $2p_0$ transition it will produce a linear superposition of $1s(A_1)$ and $2p_0$ wavefunctions a very simple wavepacket which oscillates in time as the superposition precesses around the Bloch sphere, representing the quantum mechanical state space for two-level systems. For an ensemble, all the wavepackets initially radiate in phase, and therefore strongly, to produce coherent radiation. The coherence is lost, owing to small offsets in the resonant frequencies resulting from differences in the local environment, and the radiation weakens as the dipoles dephase on a timescale given by the inverse of the $1s(A_1)$ to $2p_0$ inhomogeneous linewidth, measureable in the frequency domain using conventional continuous wave infrared spectroscopy. However, their relative phases can be restored by a subsequent laser pulse leading to a second burst of coherent radiation the photon echo which appears later by a time equal to the time difference between the initial and rephasing pulses, in precise analogy to the well-known Hahn spin echo.

978-1-4244-6346-6/10/$26.00 ©2010 IEEE

## III. Description of the Experiment

In order to provide conclusive proof of a true photon echo we need to establish both the directional property and expected timing of the phenomenon. This is now discussed.

### A. Echo Direction

We establish the directional property of the echo ($\boldsymbol{k_E} = \boldsymbol{2k_2} - \boldsymbol{k_1}$) by measuring the angular distribution of the beams. In our experiment the pump ($\boldsymbol{k_1}$) and rephasing ($\boldsymbol{k_2}$) beams intersect at an angle of -5$^o$. Simple geometry shows that the echo should emerge at an angle +5$^o$ with respect to the direction of the rephasing beam $\boldsymbol{k_2}$. Figure 1 shows the intensities of the three beams exiting the sample as a function of angle. This graph shows that the echo emerges at the predicted angle with respect to the direction of the rephasing beam $\boldsymbol{k_2}$.

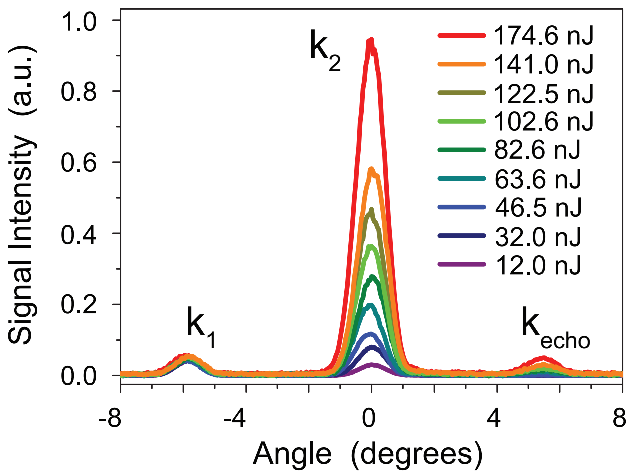


Fig. 1. Angle resolved echo. The intensities of the angle resolved signals were recorded by translating the detector across the far-field which shows that ($\boldsymbol{k_E} = \boldsymbol{2k_2} - \boldsymbol{k_1}$) as predicted.

### B. Echo Timing

Second we consider the echo arrival time, which we determine using a reference pulse split from the rephasing pulse and a delay line. The transmitted pump, rephasing and emitted echo pulses, as well as the reference pulse are all focussed onto the detector through a pinhole to produce a characteristic interference pattern in time. We exploit the angular dispersion of the pump, rephasing and echo pulses and block all but one of them, thereby obtaining the interference patterns of the reference beam with the pump, rephasing and echo beams separately. By subtracting the mean intensity and squaring the result, the arrival times and shapes of the pump, rephasing and echo pulses can then be determined as a function of time (Figure 2). All three pulses take the form of well-defined peaks, with the maxima occurring at the times anticipated for echoes.

Figure 3 shows a plot of the results of several time resolved cross-correlation experiments, similar to those shown in figure 2. A range of pump-rephasing delays $\tau_{12}$, were used to

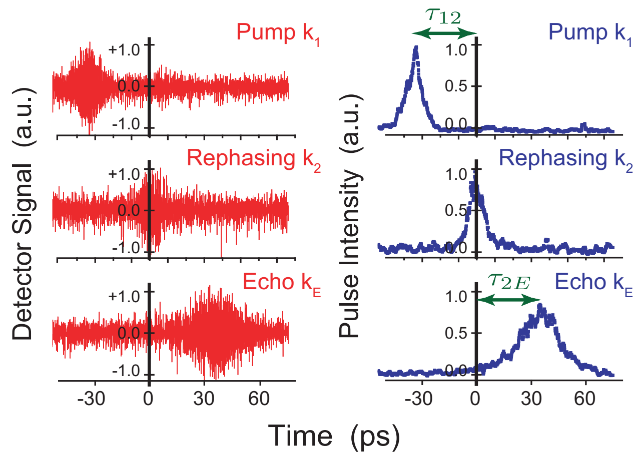


Fig. 2. Time resolved echo. On the left is the detector signal showing the interference patterns with the pump, rephasing and echo beams. A moving average has been subtracted, in order to remove the background and laser drift. The pump, rephasing and echo temporal profiles were obtained from the square of these interference patterns, as shown on the right, where the pump rephasing beam time interval $\tau_{12}$ and the rephasing beam-echo time interval $\tau_{2E}$ are also shown.

establish values of the echo delay with respect to the rephasing beam, $\tau_{2E}$. This figure shows that within experimental error, the echo arrives when expected, a finding which is independent of peak pump areas $A_P$.

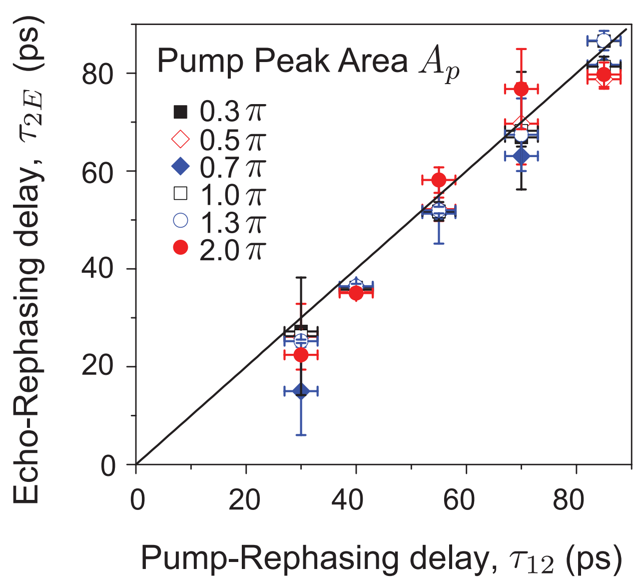


Fig. 3. Echo arrival time control. Time resolved cross-correlation experiments, similar to those shown in figure 2, with a range of pump-rephasing delays $\tau_{12}$, were used to establish values of the echo delay with respect to the rephasing beam, $\tau_{2E}$. Within experimental error, the echo arrives when expected, a finding which is independent of peak pump areas $A_P$

### C. Rabi Oscillations

We now go further to exploit the photon echo as an experimental tool to investigate the quantum coherence properties of the excited donor states. We use the photon echo to directly observe Rabi oscillations produced by coherent optical excitation of phosphorus donors in silicon with intense THz pulses from the free-electron laser. Figure 4 shows the time-integrated photon echo signal $S$ as a function of pump peak pulse area $A_P$ for a rephasing peak pulse area of $0.54\pi$ and a pulse length of 6.79 ps. The dotted line is the ideal theoretical result showing Rabi oscillations. The black line shows the corrected prediction when including the non-uniform spatial profile of the laser beam, and the magenta line includes the effect of both photoionization and the beam profile. The theory lines were calculated using values for $\mu_{12}$, $\Gamma_0$, $\sigma_{2p0}$ and $\sigma_e$ that were found from a global fit of many experimental data sets like the one shown here. The experimental results for the same conditions are shown as points. The normalisation factor for the ordinate of the experiment relative to the theory was found by a global comparison of many similar experiments with different pulse lengths and rephasing pulse areas. The error bars shown indicate the standard deviation of the normalisation factor (systematic for an individual experiment such the one in this figure) and dominate the statistical errors of the measurements.

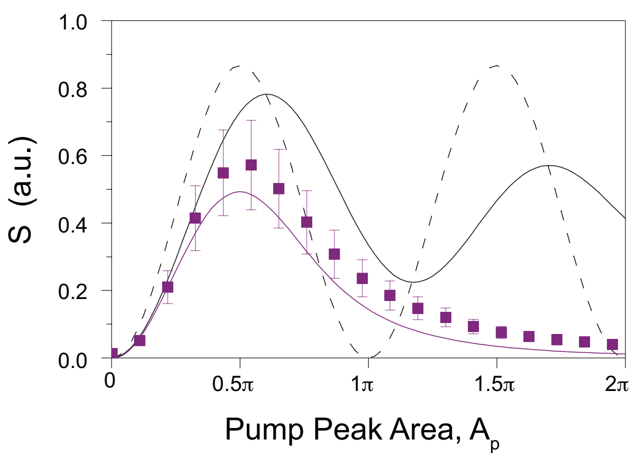


Fig. 4. The time-integrated photon echo signal S as a function of pump peak pulse area $A_P$ for a rephasing peak pulse area of $0.54\pi$ and a pulse length of 6.79 ps. The dotted line is the ideal theoretical result showing Rabi oscillations. The black line shows the corrected prediction when including the non-uniform spatial profile of the laser beam, and the magenta line includes the effect of both photoionization and the beam profile. The experimental results for the same conditions are shown as points. The error bars shown indicate the standard deviation of the normalisation factor (systematic for an individual experiment such the one in this figure) and dominate the statistical errors of the measurements.

## IV. Conclusion

We have provided the first demonstration a THz photon echo from phosphorus donors in silicon. We provide conclusive proof of this claim by establishing both the directional property and expected timing of the phenomenon. We then go further to exploit the photon echo as an experimental tool to investigate the quantum coherence properties of excited donor Rydberg states of phosphorus in silicon.

Our work shows that we can prepare coherent mixtures of different orbital states for one of the most common impurities in the most common semiconductor. Coherent control of donor orbitals in silicon opens up many possibilities such as entanglement of pairs of impurities whose ground state wavefunctions are too compact to interact. This could ultimately be exploited in a number of silicon-based quantum computing schemes that have been proposed in the literature.


## Acknowledgment

We acknowledge the financial support of NWO and EPSRC (COMPASSS, Grant Ref EP/H026622/1 and Advanced Research Fellowship EP/E061265/1).


## References


[1] B. Kane, "A silicon-based nuclear spin quantum computer," *Nature*, vol. 393, pp. 133–137, May 1998.

[2] A. Stoneham, A. Fisher, and P. Greenland, "Optically driven silicon-based quantum gates with potential for high-temperature operation," *J. Phys. Condens. Matter*, vol. 15, no. 27, p. L447, July 2003.

[3] G. Morley, D. McCamey, H. Seipel, L.-C. Brunel, J. van Tol, and C. Boehme, "Long-lived spin coherence in silicon with and electrical spin trap readout," *Phys. Rev. Lett.*, vol. 101, p. 207602, November 2008.

[4] N. Vinh, P. Greenland, K. Litvinenko, B. Redlich, A. van der Meer, S. Lynch, M. Warner, A. Stoneham, G. Aeppli, D. Paul, C. Pidgeon, and B. Murdin, "Silicon as a model ion trap: time domain measurements of donor Rydberg states," *Proc. Natl. Acad. Sci. USA*, vol. 105, no. 31, pp. 10 649–10 653, August 2008.

[5] S. Pavlov, R. Zhukavin, E. Orlova, V. Shastin, A. Kirsanov, H.-W. Hübers, K. Auen, and H. Riemann, "Stimulated emission from donor transitions in silicon," *Phys. Rev. Lett.*, vol. 84, no. 22, pp. 5220–5223, May 2000.

[6] S. Pavlov, H.-W. Hübers, M. Rümmeli, R. Zhukavin, E. Orlova, V. Shastin, and H. Riemann, "Far-infrared stimulated emission from optically excited bismuth donors in silicon," *Appl. Phys. Lett.*, vol. 80, no. 25, pp. 4717–4719, June 2002.

[7] S. Pavlov, H.-W. Hübers, H. Riemann, R. Zhukavin, E. Orlova, and V. Shastin, "Terahertz optically pumped si:sb laser," *J. Appl. Phys.*, vol. 92, no. 10, pp. 5632–5634, November 2002.

[8] H. Hübers, S. Pavlov, H. Riemann, N. Abrosimov, R. Zhukavin, and V. Shastin, "Stimulated terahertz emission from arsenic donors in silicon," *Appl. Phys. Lett.*, vol. 84, no. 18, pp. 3600–3602, May 2004.

[9] S. Lynch, P. Townsend, G. Matmon, D. Paul, M. Bain, H. Gamble, J. Zhang, Z. Ikonic, R. Kelsall, and P. Harrison, "Temperature dependence of terahertz optical transitions from boron and phosphorus dopant impurities in silicon," *Appl. Phys. Lett.*, vol. 87, p. 101114, September 2005.

[10] S. Lynch, D. Paul, P. Townsend, G. Matmon, Z. Suet, R. Kelsall, Z. Ikonic, P. Harrison, J. Zhang, D. Norris, A. Cullis, C. Pidgeon, P. Murzyn, B. Murdin, M. Bain, H. Gamble, M. Zhao, and W.-X. Ni, "Towards silicon-based lasers for terahertz sources," *IEEE J. Select. Topics. Quantum Electron.*, vol. 12, no. 6, pp. 1570–1578, November/December 2006.

[11] S. Pavlov, U. Bottger, N. Abrosimov, K. Irmscher, H. Riemann, and H.-W. Hübers, "Influence of an electric field on the operation of terahertz intracenter silicon lasers," *J. Appl. Phys.*, vol. 107, no. 3, p. 033114, February 2010.

[12] P. Greenland, S. Lynch, A. van der Meer, B. Murdin, C. Pidgeon, B. Redlich, N. Vinh, and G. Aeppli, "Coherent control of Rydberg states in silicon," *accepted Nature*, April 2010.

[13] C. Jagannath, Z. Grabowski, and A. Ramdas, "Linewidths of the electronic spectra of donors in silicon," *Phys. Rev. B*, vol. 23, no. 5, pp. 2082–2098, March 1981.

[14] D. Karaiskaj, J. Stotz, T. Meyer, M. Thewalt, and M. Cardona, "Impurity absorption spectroscopy in $^{28}$Si: the importance of inhomogeneous isotope broadening," *Phys. Rev. Lett.*, vol. 90, no. 18, pp. 186 402–1, May 2003.