

Lifetime Measurements of Group V Donor Rydberg States in Silicon at THz Frequencies

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Abstract—We have measured the T_1 lifetimes of Rydberg states of phosphorus and arsenic in silicon at THz frequencies using the FELIX pulsed free electron laser. Our results show the dominant decoherence mechanism is lifetime broadening.

I. INTRODUCTION

Group V donors in silicon have recently been attracting considerable attention. They have been shown to exhibit lasing at THz frequencies under mid-IR optical pumping [1], [2] and have the potential for electrically pumped lasing [3], [4]. It has also been suggested that group V donors could form the basis of an optically controllable Qubit in a silicon-based quantum computer [5]. We have recently shown that silicon doped with group V donors has many attractive features for this important application [6]. In this paper, we discuss time-domain lifetime measurements of Rydberg states of phosphorus and arsenic in silicon at THz frequencies using the FELIX pulsed free electron laser. Our results show that the dominant decoherence mechanism is lifetime broadening.

II. BACKGROUND TO THE EXPERIMENT

Donor atoms in Si behave like a hydrogen atoms embedded in a dielectric matrix. The difference, however, is that the mass of the donor atom nucleus is now so much heavier than the mass of the proton that it can be assumed infinite, and the Coulomb attraction between the electron and donor atom nucleus is much weaker because it is strongly screened. The net result is that the Rydberg is now shifted to much smaller energies (50 meV for phosphorus in silicon as apposed to 13.6 eV for hydrogen). The resulting Lyman series is therefore shifted to THz frequencies as depicted in Fig. 1.

The spectroscopy of excited Rydberg states of donors in silicon has a long and distinguished history, and remains an active field of research today, with particular attention given to the extraordinarily narrow linewidths of some of the transitions. In the limit of a very clean, homogeneous material, frequency domain spectroscopy provides direct information about the relaxation dynamics. For a real material, however, determination of relaxation times from the frequency domain linewidth is notoriously difficult because the observed shape of the absorption line is generally given by a convolution of

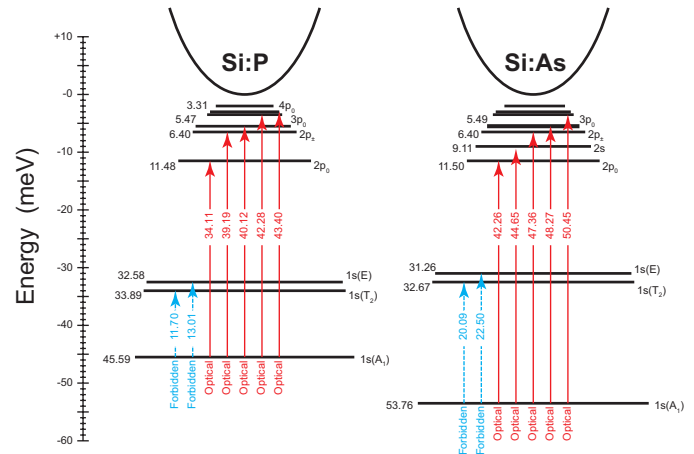


Fig. 1. Rydberg series and optical transitions from the lowest energy states of the hydrogen-like donor impurities phosphorus and arsenic in silicon.

the homogenous (or natural) linewidth with the instrument response and a variety of inhomogenous broadening mechanisms. The latter include random strain fields induced by impurities and/or dislocations, and other fluctuations in the donor environment due to chemical impurities and different isotopes in the natural composition of Si with differing nuclear moment. We report the first direct time domain measurement of T_1 . We have used the free electron laser FELIX to perform these measurements because the required laser power, pulse duration, frequency coverage, and duty cycle are ideally matched to this application. FELIX gives continuous coverage over the spectral range 5 to 400 meV, controllable pulse durations of between 6 and 100 optical cycles, and peak powers up to 100 MW.

III. EXPERIMENTAL DETAILS AND RESULTS

The experiment was performed using a three-beam degenerate pump-probe technique described in detail elsewhere [7]. Fig. 2 shows the measured probe transmission change for the $1s(A_1) \rightarrow 2p_0$ transition at 34.1 meV in phosphorus doped silicon. The rise of the leading edge indicates the pulse duration, which was 10 ps in this case. For all pump

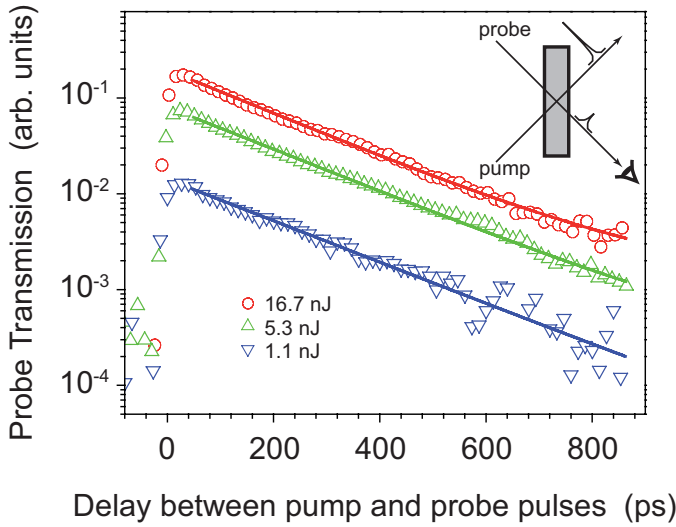


Fig. 2. The change in probe transmission induced by the pump as a function of the time delay between pump and probe, observed in the Si:P sample for the $1s(A_1) \rightarrow 2p_0$ transition at a sample temperature, T , of 10 K and a pump and probe photon energy of 34.1 meV. The rise of the leading edge indicates the pulse duration, which was 10 ps. The laser pump powers used corresponded to micropulse energies shown on the figure. The lowest pump pulse energy (1.1 nJ) corresponds to a focussed photon fluence of $1017 \text{ photons m}^{-2}$. Also shown are fits using a single exponential decay where the decay parameter is the spontaneous relaxation rate $1/T_1$. The inset shows the transient pump-probe experimental geometry.

powers the decay is (almost) exponential; the signal is (almost) linear when plotted on a log-linear scale. Numerical fits with a simple exponential decay gave a value for the lifetime of $T_1 = 205 \pm 18 \text{ ps}$. This corresponds to a linewidth of $1/T_1 = 0.026 \text{ cm}^{-1}$, i.e. less, but not very much less than the lowest value reported for this transition of 0.034 cm^{-1} , which was obtained in an isotopically pure ^{28}Si sample [8]. Fig. 3 shows the absorption spectrum and lifetimes for P doped Si and As doped Si samples. The absorption spectrum was measured with a resolution of 0.25 cm^{-1} (0.03 meV) at 10 K. The well-known Lyman transitions $1s(A_1) \rightarrow np_0, np_{\pm}$ between 34 and 45 meV are apparent. No pump-probe signal was seen when the laser was not resonant. The relaxation lifetime of the $2p_0$ state in Si:P has the longest lifetime since it is farthest from the peak in the density of phonon states. The fact that the $2p_{\pm}$ shows a slightly shorter lifetime than the $2p_0$ is also consistent with the spectroscopic linewidth [8]. The lifetime of the $2p_0$ state in Si:As is slightly shorter than that of Si:P but we note that in spite of this it is potentially more useful because the energy gap between Rydberg states is larger and overlaps with the available wavelength range of far-infrared semiconductor diode laser pump sources.

In order to fully understand our results we developed a simple 3-rate-equation model. This took account of excitation and recombination between the ground level, bound Rydberg state, and the continuum of states just above the conduction band edge. By fitting the data using realistic physical and experimental parameters, we were able to determine unambiguously that we had measured the true homogenous lifetime.

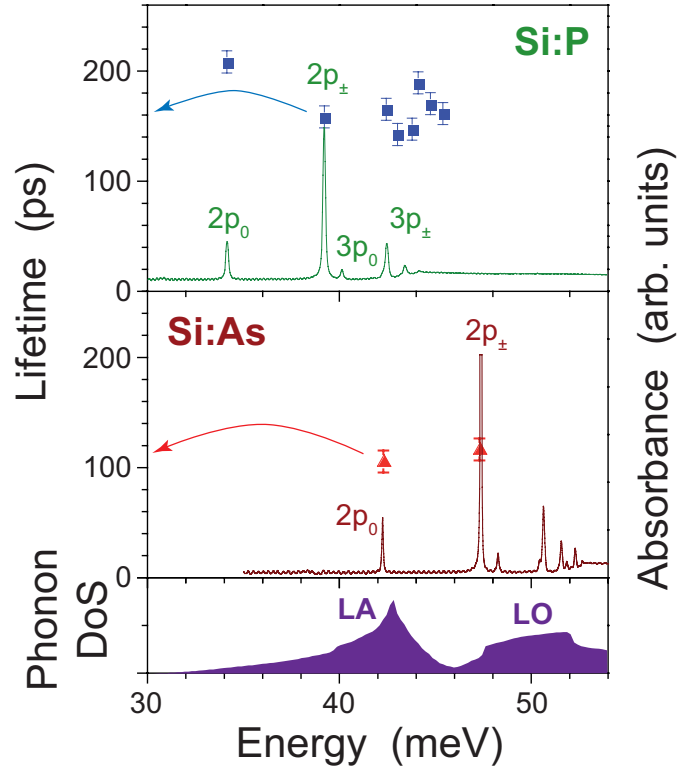


Fig. 3. (a) The absorption spectrum for P-doped Si measured by FTIR spectroscopy with 0.25 cm^{-1} or 0.03 meV resolution. The sample temperature was 10 K. The lifetimes of the indicated states, determined from pump-probe signals (such as those in Fig. 2) are also shown. (b) The corresponding results for Si:As. (c) The one-phonon density of states of silicon, which, of course, determines the phonon emission decay rate at low temperature

We were also able to demonstrate that other possible effects such as multi-photon ionisation play no significant role in the measurement by fitting the data using our model. An example is shown in Fig. 4. Close inspection of the rate equations reveals that relaxation from the excited Rydberg state to the ground level should give rise to a purely exponential decay. If multi-photon ionisation to the continuum plays a significant role then we would expect the decay to be dominated by a $1/T$ dependence. Fig. 4 demonstrates unequivocally that multi-photon ionization was negligible in our measurement.

IV. TEMPERATURE DEPENDENCE

The lifetime values referred to in the previous section were all measured at 4.2 K. For completeness we also measured the temperature dependence of the $2p_0$ and $2p_{\pm} \rightarrow 1s(A_1)$ decay times. These decay times are shown in Fig. 5. This figure shows the remarkable result that the decay times actually increase with temperature - more obviously for $2p_{\pm}$ than for $2p_0$ - displaying a maximum at 50 K. We are able to explain this temperature dependence with a phenomenological equation for the effective relaxation time based on [9]. Using this equation (see inset of Fig. 5 for a key to the symbols) we were able to fit this data and extract the following values, $T_1 = 215 \pm 10 \text{ ps}$, $\Delta E_a = 11.8 \pm 1.1 \text{ meV}$, $\Delta E_b = 32.2 \pm 2.1 \text{ meV}$,

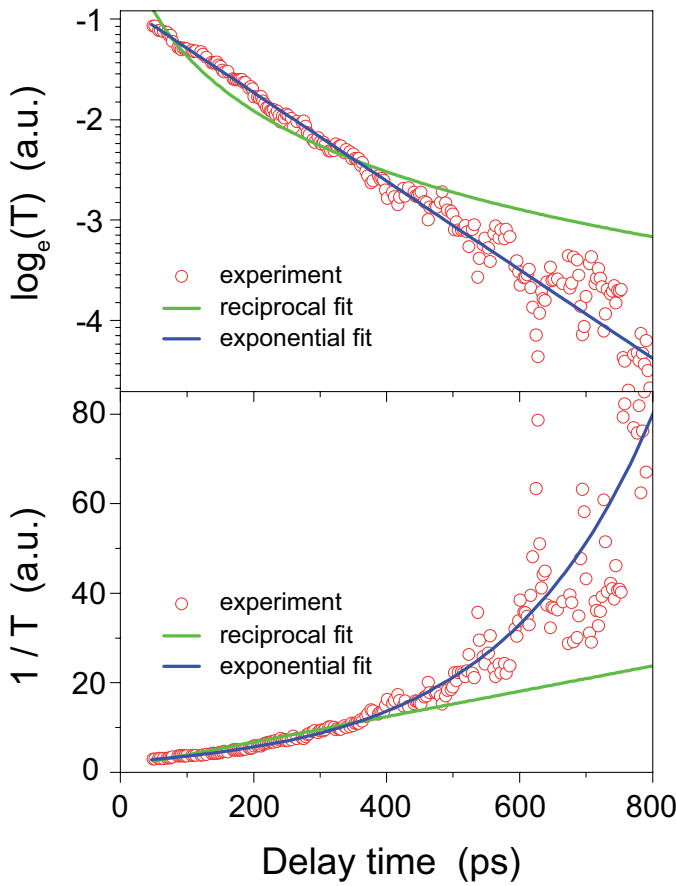


Fig. 4. The probe transmission at high pump intensity is shown as a function of probe delay, along with a fitted single exponential decay and a reciprocal decay. We plot both the logarithm (upper panel) and reciprocal (lower panel) of the signal. If spontaneous decay is most important we expect the former to be linear as a function of time; if recombination from the conduction band is most important, then the latter will show linear behaviour. At high intensities, even though two-photon ionization is likely to be strong, the experimental signal is exponential.

and $1/R_{TO} = 1.7 \text{ ps}$ from the fitting parameters, in good agreement with literature values.

V. CONCLUSION

We have measured the lifetime of excited donor Rydberg states directly by a time-domain method. Comparing with literature values for lifetimes from the frequency domain, we find that, at low temperature the dominant decoherence mechanism is *lifetime* broadening due to the emission of phonons. In addition, we show that there is a remarkable insensitivity of the results to multi-photo ionisation effects as the power of the intense free electron beam is varied, and discover that initially the recovery time for the $2p \rightarrow 1s$ absorption actually increases with temperature.

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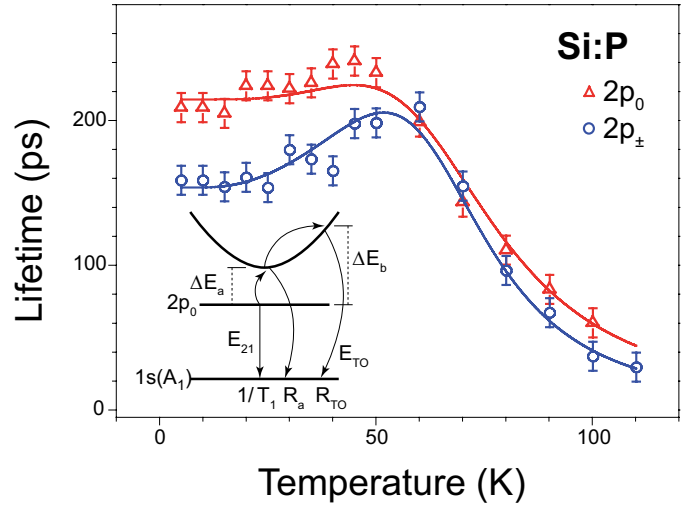


Fig. 5. The temperature dependence of the lifetimes for $1s(A_1) \rightarrow 2p_0, 2p_{\pm}$ transitions at 34.1 and 39.2 meV, respectively in Si:P. The lines are fits of the phenomenological our equation. The inset shows the level scheme with transitions.

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