Optical Detection and Ionization of Donors in Specific Electronic and Nuclear Spin States

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We resolve the remarkably sharp bound exciton transitions of highly enriched ^{28}Si using a single-frequency laser and photoluminescence excitation spectroscopy, as well as photocurrent spectroscopy. Well-resolved doublets in the spectrum of the ^{31}P donor reflect the hyperfine coupling of the electronic and nuclear donor spins. The optical detection of the nuclear spin state, and selective pumping and ionization of donors in specific electronic and nuclear spin states, suggests a number of new possibilities which could be useful for the realization of silicon-based quantum computers.

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The proposal [1] of a scaleable quantum computer based on the $I = 1/2$ nuclear spin of ^{31}P donors in a spin-free ^{28}Si host has led to many ingenious suggestions and modifications [2–9] seeking to take advantage of the highly developed state of silicon materials science and nanofabrication technology. The isolation from the environment responsible for the record [10] coherence times of nuclear spins in semiconductors brings with it the inevitable cost of difficulty in determining the quantum state of the qubit. Optical measurement and control of nuclear spins via the hyperfine interaction, important in approaches using trapped ions or atoms, is absent from virtually all solid-state proposals, since these splittings are thought to be beyond the resolution limits of solid-state spectroscopy, except for the special case of rare earth ions in some insulating hosts [11,12]. Recent studies [13] of shallow bound exciton (BE) photoluminescence (PL) in highly enriched ^{28}Si revealed a remarkable reduction in linewidth compared to natural silicon, leading to a suggestion [14] that it might be possible to determine the nuclear spin of an isolated ^{31}P impurity in ^{28}Si by detecting the donor hyperfine splitting in the phosphorus BE transition. We show that the situation is much more favorable than suggested by the early results [13], using PL excitation spectroscopy (PLE) with neV resolution to demonstrate well-resolved hyperfine splittings in the BE Zeeman spectrum of an ensemble of ^{31}P donors in highly enriched ^{28}Si. The hyperfine splittings are also observed in photocurrent spectroscopy, demonstrating efficient selective photoionization of ^{31}P in specific electronic and nuclear spin states via BE Auger recombination. This leads us to propose a novel readout mechanism for single impurity nuclear spin states—the optical nuclear spin transistor.

Great progress has been made in the manipulation and measurement of qubits based on electronic spin states in semiconductors, including the electrical detection of the spin resonance of a single defect [15], the detection of a single spin by magnetic resonance force microscopy [16], the single-shot electrical readout of an electron spin in a quantum dot [17], and the optical detection and manipulation of the electronic spin states of isolated nitrogen-vacancy (NV) centers in diamond, including controlled coupling to the electronic and nuclear spins of other nearby centers [18–21]. The use of the nuclear spins of impurities in semiconductors as qubits, while offering the advantage of very long coherence times, lacks demonstrated mechanisms for reading out single spins (one approach circumvents this problem by having many identical copies of a given qubit, so the signal becomes large enough to be read out with existing methods [3,7]). The proposed [14] direct optical measurement of the nuclear spin state of a single ^{31}P impurity in ^{28}Si is therefore an exciting possibility.

It should be emphasized that even in natural Si (92.23% ^{28}Si, 4.67% ^{29}Si, 3.10% ^{30}Si), the 5 μeV FWHM of the phosphorus BE no-phonon PL transition was among the narrowest near-gap transitions known in semiconductors [13]. The discovery that it narrowed to the instrumental resolution limit in highly enriched ^{28}Si was the first indication of the importance of inhomogeneous isotope broadening in determining the observed linewidth of this and other electronic transitions in Si [13,22]. The resolution limit imposed by the use of commercial spectrometers, together with the weak Si PL signal, has been overcome by

studying the no-phonon transitions of the BE in absorption rather than in emission, using a tuneable single-frequency laser source with a linewidth of less than 0.3 neV, and detecting the weak absorption by measuring the resulting transverse optical (TO) phonon replica luminescence signal. An improved sample of ^{28}Si was also available for the present study, with an isotopic enrichment of 99.991%, and much higher chemical purity, having phosphorus $\sim 2 \times 10^{12}$ cm^{-3} and boron $\sim 5 \times 10^{13}$ cm^{-3}, as measured by photoluminescence, and carbon $< 5 \times 10^{14}$ cm^{-3} (detection limit) as measured by local vibrational mode absorption.

The distributed feedback Yb-doped fiber laser could be temperature tuned over the region of interest, and the laser frequency was monitored to one part in 10^7 with a wave meter. The laser output was amplified to 500 mW in an Yb-doped fibre amplifier, mechanically chopped to allow for lock-in detection, and focused onto the edge of the sample. Samples were loosely mounted (to avoid strain) in a reflecting cavity to optimize the weak signals, and immersed in liquid He. Additional above-gap excitation to photoneutralize the impurities was provided by a 1184 meV (1047 nm) Nd:YLF laser at power levels between 1 and 500 mW. Note that while these optical power levels appear high, both laser beams are very weakly absorbed in the sample. The PL signal was separated from the scattered excitation radiation using a 3/4 m double monochromator and detected with a Ge photoconductive detector. The resolution of the double monochromator was far less than what would be required to separate the BE components studied here, so that the PLE detection was nonselective. The pump/luminescence beams entered or exited the sample perpendicular to the magnetic field direction, but due to the high transparency and refractive index of Si, and the high collection efficiency geometry employed here, multiple reflections would tend to reduce any direction and polarization effects.

Figure 1(a) shows a typical PLE spectrum of the phosphorus BE no-phonon transitions for the new sample in the presence of a small magnetic field, revealing six well-resolved hyperfine-split doublets. The spectrum of the same transitions in silicon having the natural isotopic composition is also shown, making it clear why the observation of hyperfine splittings in such transitions had been thought to be impossible. Figure 1(b) shows a level diagram for the observed transitions between the magnetically split levels of the neutral donor, D^0, and the neutral donor BE, D^0X. Note that while the Zeeman splitting of the electron in D^0 and of the hole in D^0X increase linearly with magnetic field, the donor hyperfine splittings become constant at intermediate fields, and their sum equals the zero-field splitting. At the fields and temperatures used here, all of the splittings are much less than kT. The electron and hole g factors determined from these spectra, $g_e = 1.97$, $g_{h1/2} = 0.83$ and $g_{h3/2} = 1.3$ are in good agreement with earlier studies [23] of the phosphorus BE

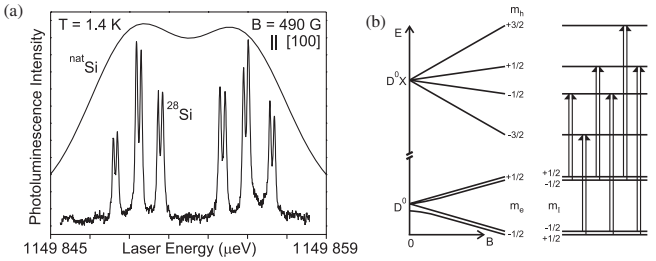

FIG. 1. Zeeman spectrum of the phosphorus bound exciton no-phonon transitions in a sample enriched to 99.991% ^{28}Si. (a) The PLE spectrum at $T = 1.4$ K with a 490 G field parallel to the [100] axis. For comparison, the spectrum of natural Si is shown, shifted to compensate for the dependence of the band gap energy on the average isotopic mass [13,22]. (b) Level scheme showing the origin of the transitions. D^0 is the ground state of the phosphorus neutral donor, which has a zero-field hyperfine splitting of 486 neV, and under an applied field splits into four hyperfine levels determined by the projections of the electron spin $m_e = \pm\frac{1}{2}$ and the nuclear spin $m_I = \pm\frac{1}{2}$. The donor bound exciton D^0X has two electrons in a spin singlet, and under a magnetic field splits only according to the $j = 3/2$ hole ''spin'' projection, m_h. The six doublet transitions are ordered in increasing energy from left to right, in correspondence with the transitions shown in (a). The two ''forbidden'' transitions with $\Delta m = \pm 2$ are not shown.

at much higher fields, and the sum of the hyperfine doublet splittings agrees with the 486 neV (117.53 MHz) phosphorus donor hyperfine splitting determined by EPR [24].

Figure 1(a) demonstrates convincingly that the donor hyperfine splittings can be well resolved in the BE transitions in highly enriched ^{28}Si even for an ensemble of donors. A fit to the 12 components gives an average FWHM of 150 neV, average hyperfine splittings of 227 (245) neV for the higher (lower) energy D^0 Zeeman level, and a selectivity of 25 for one hyperfine state over the other when pumping at the peak of a subcomponent. Even better results can be expected for a single donor, when only the homogeneous linewidth is important. The 272 ns lifetime [25] of the phosphorus BE sets a lower limit of ~ 5 neV on the homogeneous linewidth. While this may in the future be directly measurable, we next show that ensemble linewidths considerably less than those demonstrated here should be achievable in ^{28}Si samples having higher enrichment. In Fig. 2 one of the hyperfine doublets shown in Fig. 1(a) is compared with that of the same sample at 4.2 K, as well as those of two other samples having lower enrichment. The increase in temperature from 1.4 to 4.2 K produces only a 27% increase in linewidth, indicating that while temperature does play a role, it is not a major contributor to the low temperature linewidth. As compared to the well-resolved splittings for the sample enriched to 99.991%, the hyperfine splitting is barely resolved for the sample with an enrichment of 99.983%, and is not resolved for the sample with 99.92% ^{28}Si.

These differences can be explained by a simple argument which assumes that inhomogeneous isotope broad-

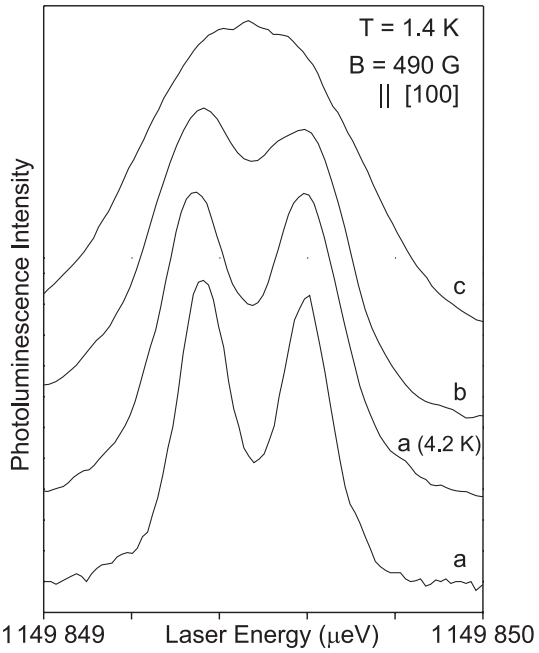

FIG. 2. Dependence of the phosphorus bound exciton PLE line shapes on temperature and isotopic enrichment. At the bottom is a section of the spectrum shown in Fig. 1(a), while above it is a spectrum of the same sample at a temperature of 4.2 K (shifted up in energy to compensate for the temperature dependence of the band gap energy [27]). The spectrum labeled b is for a sample enriched to 99.983% ^{28}Si, and the spectrum labeled c is for a sample enriched to 99.92% ^{28}Si, both at 1.4 K.

ening remains the dominant mechanism even at the highest enrichment studied here. Ignoring for simplicity the fact that the ratio of ^{29}Si to ^{30}Si may change between natural Si and the enriched samples, the inhomogeneous isotope broadening should vary as the square root of the ^{29}Si + ^{30}Si content. The 5 μeV FWHM for natural Si scales by this simple argument to widths of 510, 230, and 170 neV for the samples shown in Fig. 2, in order of increasing enrichment, in remarkably good agreement with what is observed. Narrower ensemble linewidths should therefore be achievable in samples with higher enrichment.

As is often the case in semiconductor spectroscopy, the interpretation of the PLE spectrum in the absence of an external magnetic field is more challenging than when a uniform field is applied, since the external perturbation can lift degeneracies and dominate over the effects of random interimpurity interactions. Figure 3 shows a typical zero-field spectrum of the same sample shown in Fig. 1(a). The larger bracket at the bottom indicates the zero-field hyperfine splitting of 486 neV, while the two smaller brackets indicate an additional splitting which varies with sample impurity content, temperature, and excitation conditions. Note that since the 99.991% ^{28}Si sample is p type due to the dominant boron impurity, at 1.4 K there is virtually no resonant PLE signal from the phosphorus BE unless the sample is also weakly excited with above-gap light to generate free electrons and holes and thus photoneutralize

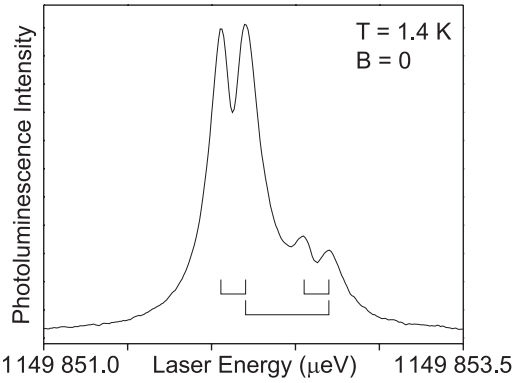

FIG. 3. Photoluminescence excitation spectrum of the zero-field structure of the phosphorus bound exciton transition in the sample enriched to 99.991% ^{28}Si. The large bracket at the bottom indicates the 486 neV zero-field hyperfine splitting of the phosphorus donor, while the two smaller brackets label an additional splitting which varies with experimental conditions.

the ionized donors. The intensity of this nonresonant excitation has a strong effect on the additional zero-field splitting seen in Fig. 3, although it does not affect the splittings shown in Fig. 1(a). Ideally one would use a sample containing only phosphorus and no acceptors, eliminating the need for photoneutralization. The additional splittings observed at zero field likely arise from interactions between the spin of the donor electron and that of other nearby impurities, whose charge states are controlled by the above-gap excitation. A more complete description of the zero-field behavior will be given in a following, comprehensive publication.

Shallow donor and acceptor BE in Si and other indirect band gap semiconductors have very low radiative quantum efficiencies due to the dominance of nonradiative Auger recombination [25], which is a drawback for optical readout of the impurity spin state [14]. For the phosphorus BE, the observed 272 ns lifetime is essentially equal to the Auger lifetime, since the radiative lifetime is $\sim$2 ms [25]. However, it is possible to put the dominant Auger recombination to use by detecting the free electrons released in the Auger process. In Fig. 4 we see the spectrum of the phosphorus BE under conditions similar to those used in Fig. 1(a), revealed by photocurrent spectroscopy rather than by PLE. Simple electrical contacts were made to the ends of the 2.5 cm long sample by rubbing on a thin layer of In-Ga eutectic, using fine copper wires to connect an external 1.5 V bias source and a transimpedance current amplifier. Strains generated by these contacts are likely responsible for the reduced resolution of Fig. 4 as compared to Fig. 1(a), but the hyperfine splittings are still clearly resolved.

The near-unity efficiency of the Auger process for these BE suggests a novel mechanism for reading out the nuclear spin of individual ^{31}P impurities—the optical nuclear spin transistor. Here a tuneable laser is used to address one or the other hyperfine state of an individual neutral ^{31}P donor,

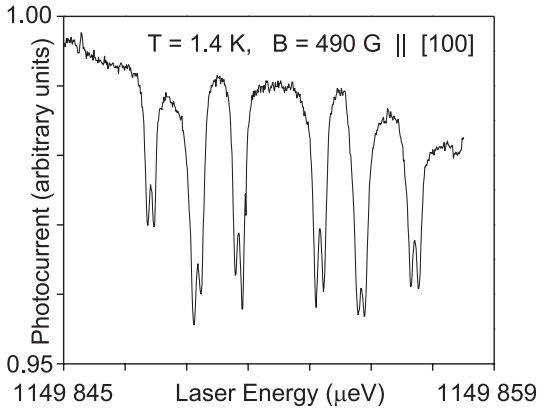

FIG. 4. Photocurrent spectroscopy of the phosphorus bound exciton transitions in a sample enriched to 99.991% ^{28}Si with a magnetic field of 490 G applied parallel to the [100] axis. Because of electron-hole recombination, the electrons from the bound exciton Auger recombination reduce the larger nonresonant hole photocurrent from the dominant impurity, boron.

creating a BE only if the donor is in the selected nuclear spin state (the donor can be maintained in the lowest electronic spin state by the use of high field and low temperature). The resulting ionization of the donor can be sensed by a nearby field effect [15] or single-electron [4–6] transistor, and the donor can then be neutralized either by the random capture of a free electron or from an electron reservoir. Individual donors can be made distinguishable by using a magnetic field gradient [3,7] or by taking advantage of inhomogeneous broadening [11,26].

The ability to optically pump and ionize donors in specific electronic and nuclear spin states also suggests that substantial nuclear polarizations should be achievable, an essential requirement for initializing the qubits before any computation. Indeed, the hyperfine subcomponents of given transitions in Fig. 1(a) and 4 are clearly not of equal intensity, indicating that some dynamical polarization of the nuclear spins is taking place (all of the splitting energies are much less than kT, so that thermal polarizations are negligible). Achieving high nuclear polarizations may require the use of *n*-type samples, so that nonresonant excitation to achieve donor photoneutralization is not required. The use of higher magnetic fields to polarize the spins of both donor and free electrons may also be beneficial. The large difference in nuclear spin flip energy between the neutral donor and the ionized donor (or donor BE) could be put to advantage, using an NMR pulse in resonance with the nuclear transitions of the ionized donor to flip the nuclear spins of only those donors which are selectively ionized by pumping one of the BE hyperfine transitions.

We have demonstrated the direct optical readout of the nuclear spin of ^{31}P impurities in ^{28}Si, and the selective ionization of donors in specific electronic and nuclear spin states, using the hyperfine splittings of the donor BE transitions. Many other optically active impurities and defects are known in Si, with widely different physical properties. For example, the high radiative quantum efficiency of isoelectronic BE may offer advantages in future quantum computing and information processing applications. Many of these centers may also reveal resolved hyperfine splittings once inhomogeneous broadening mechanisms are sufficiently reduced, and the properties of promising centers should therefore be reexamined in highly enriched ^{28}Si.

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