

## Spin-dependent processes at the crystalline Si-SiO<sub>2</sub> interface at high magnetic fields

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(Received 24 May 2008; published 2 July 2008)

An experimental study on the nature of spin-dependent excess charge-carrier transitions at the interface between (111)-oriented phosphorous-doped ( $[P] \approx 10^{15} \text{ cm}^{-3}$ ) crystalline silicon and silicon dioxide at high magnetic field ( $B_0 \approx 8.5 \text{ T}$ ) is presented. Electrically detected magnetic-resonance (EDMR) spectra of the hyperfine split  $^{31}\text{P}$  donor-electron transitions and paramagnetic interface defects were conducted at temperatures in the range of  $3 \text{ K} \leq T \leq 12 \text{ K}$ . The results at these previously unattained (for EDMR) magnetic-field strengths reveal the dominance of spin-dependent processes that differ from the previously well investigated recombination between the  $^{31}\text{P}$  donor and the  $P_b$  state, which dominates at low magnetic fields. While magnetic resonant current responses due to  $^{31}\text{P}$  and  $P_b$  states are still present, they do not correlate and only the  $P_b$  contribution can be associated with an interface process due to spin-dependent tunneling between energetically and physically adjacent  $P_b$  states. This work provides an experimental demonstration of spin-dependent tunneling between physically adjacent and identical electronic states as proposed by Kane [Nature (London) **393**, 133 (1998)] for readout of donor qubits.

DOI: [10.1103/PhysRevB.78.045303](https://doi.org/10.1103/PhysRevB.78.045303)

PACS number(s): 76.30.-v, 71.55.Cn, 73.40.Qv

### I. INTRODUCTION

Phosphorus-doped crystalline silicon (*c*-Si:P) is one of the most widely utilized semiconductor materials, with applications ranging from conventional microelectronics<sup>1</sup> to proposed and presently widely investigated concepts for spintronics<sup>2</sup> and spin-based quantum information processing (QIP).<sup>3</sup> Silicon-based spin-QIP and spintronics concepts aim to utilize the comparatively weak spin-orbit coupling present in this material, and the correspondingly very long spin-coherence times,<sup>2,3</sup> as well as the impact of spin-selection rules on electronic transitions which can be used for spin readout.<sup>4</sup> Most of these applications involve electrical transport and spin manipulation at or near the silicon-silicon dioxide (SiO<sub>2</sub>) interface, making the understanding of spin processes in this region extremely important. Numerous studies of spin-dependent transport and recombination at the interface between *c*-Si:P and SiO<sub>2</sub> have recently been undertaken with the aim of identifying and understanding these mechanisms,<sup>5-7</sup> and showing that they can be utilized for the observation of very small ensembles of donors<sup>8</sup> and coherent spin motion.<sup>7,9</sup> Additionally, spin-dependent transport in two-dimensional electron gases at the *c*-Si/SiO<sub>2</sub> interface has been demonstrated.<sup>10,11</sup> However, no systematic study of such processes at high magnetic field has been conducted to date with the only data at magnetic fields  $B_0 > 400 \text{ mT}$  given by a single electrically detected magnetic-resonance (EDMR) spectrum recorded at  $B_0 = 7.1 \text{ T}$  and a temperature  $T = 4 \text{ K}$  (Ref. 12).

In the following, a systematic investigation of the spin-dependent processes at the interface between *c*-Si:P and SiO<sub>2</sub> is presented for high magnetic fields ( $B_0 \approx 8.5 \text{ T}$ ) at temperatures in the range of  $3 \text{ K} \leq T \leq 12 \text{ K}$ . We show that the dominant spin-dependent recombination mechanism at

low magnetic fields (recombination between  $^{31}\text{P}$  and  $P_b$  centers) is not seen at high fields. Instead, transitions involving only  $^{31}\text{P}$  donors or  $P_b$  centers dominate the observed EDMR signals. This study focuses in particular on the nature of the  $P_b$ -only transition which has previously been observed at low magnetic fields and nominally undoped *c*-Si-SiO<sub>2</sub> interfaces,<sup>6</sup> but has, however, not been observed in the presence of  $^{31}\text{P}$  donors.

### II. EXPERIMENT

Experimentally, we used prime grade Cz-grown *c*-Si (111) with a phosphorus donor concentration  $[P] \approx 10^{15} \text{ cm}^{-3}$ . The sample was contacted by thermal evaporation of a 100 nm Al layer after a surface clean and subsequent removal of the native oxide by wet treatment with hydrofluoric acid. Following this procedure and the structuring of the sample contacts by a photolithographic lift-off procedure, a native SiO<sub>2</sub> layer was formed on the surface between the contacts due to the exposure of the sample to air at room temperature. Similarly to previous studies of spin-dependent recombination and transport at low magnetic fields,<sup>13-15</sup> we used EDMR to investigate these processes. With this technique, the photocurrent through a sample is monitored while electron-spin resonance is used to manipulate the spin of paramagnetic centers involved in spin-dependent transitions. The latter are detected by measurement of currents which change from a constant offset value under spin-resonance conditions.<sup>16</sup> In order to perform spin resonance at  $B_0 \approx 8.5 \text{ T}$ , the quasi-optical 240 GHz superheterodyne spectrometer facility at the National High Magnetic Field Laboratory in Tallahassee, Florida was used.<sup>17</sup> A sample compatible to the geometric constraints of the Fabry-Pérot (FP) resonator of the spectrometer was used for the experiments, and is shown sche-

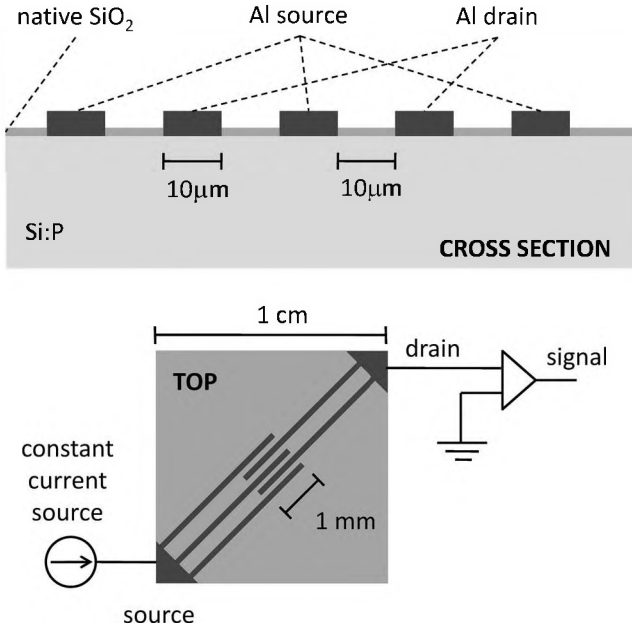


FIG. 1. A sketch of the sample used in these experiments. The sample is fabricated on Si(111) doped with  $10^{15}$  phosphorus donors/cm<sup>3</sup>. Contacts are made to the sample using aluminum contacts and the surface is covered with a native SiO<sub>2</sub> layer. A simple measurement circuit is also shown. The sketch is not to scale, although the indicated dimensions are accurate.

matically in Fig. 1. It consists of an approximately 330- $\mu$ m-thick  $8 \times 8$  mm<sup>2</sup> silicon substrate sandwiched between two 160- $\mu$ m-thick quartz slabs needed as antireflection coatings to allow the 240 GHz radiation to be coupled into the silicon bulk. The electrical contacts to the device are a 100- $\mu$ m-wide grid structure consisting of five 10- $\mu$ m-wide interdigitated fingers, with 10  $\mu$ m separation between the opposite fingers. The contact fingers were approximately 6 mm long, yet fingers belonging to the two opposite contacts overlapped by only 1 mm. This geometry ensured that: (i) The active region of the sample was located on the optical axis of the FP resonator such that the microwave field  $B_1$  was maximal and homogeneous throughout the active area; (ii) the external contacts of the sample (which were contacted with silver paste) were well outside the  $B_1$  field such that they could not distort the FP resonator modes; and (iii) due to the length of the contacts (almost 12 mm, stretching across the entire beam diameter) all metal structures within the beam diameter were aligned perpendicular to the polarization of the  $B_1$  field, reducing loss due to microwave absorption. The high magnetic field  $B_0$  is aligned normal to the sample surface. The photocurrent needed for the EDMR experiments was induced by white light (cold light) generated by a xenon discharge lamp, filtered of its infrared component and coupled into the sample via an optical fiber.

For the data acquisition, the microwave radiation was modulated which allowed a lock-in detection of the magnetic-resonance induced current changes. The relative current change  $\Delta I/I$  observed is shown in Fig. 2(a) for  $T = 3, 6,$  and  $12$  K. There are three resonances clearly visible which (as for all data presented in this study) were fit by Gaussian functions. The two resonances at the highest mag-

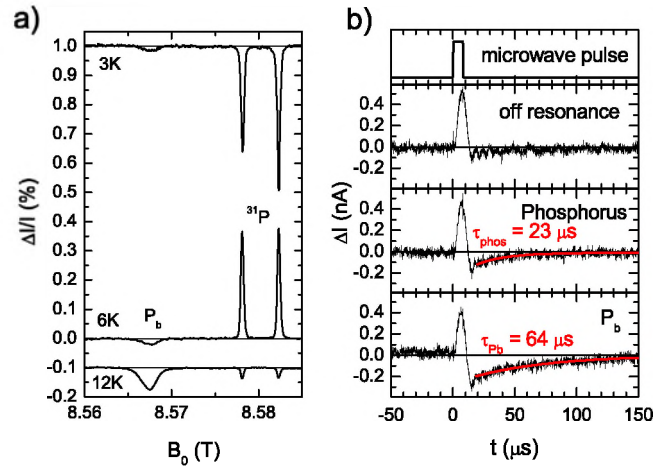


FIG. 2. (Color online) (a) Plots of the relative photocurrent changes as a function of the applied magnetic field  $B_0$  at temperatures  $T=3, 6,$  and  $12$  K. The data were taken with a photocurrent  $I=600$  nA. The signal was obtained by microwave chopping ( $500 \mu$ s pulse length, 1 kHz shot repetition rate) and lock-in detection of the photocurrent. (b) Plots of the photocurrent changes as a function of time following a microwave pulse with  $\tau_p=8 \mu$ s length for magnetic fields off resonance, on resonance with the low-field  $^{31}\text{P}$  peak, and on resonance with the  $P_b$  peak at  $T=12$  K. The data were collected from the average of many transients measured with a 1 kHz shot repetition rate.

netic fields are separated by  $B=4.2$  mT as expected from phosphorus donor electrons due to their hyperfine coupling to the donor nuclear spin. These resonances were used to calibrate the magnetic-field axis (with the applied frequency set to 240 GHz) due to the drift in the superconducting magnet and the internal field due to the polarized electrons.<sup>18</sup> The resonance at lower magnetic field at a  $g$ -factor of  $g = 2.0014$ , is assigned to the  $P_b$  interface defect (a silicon dangling bond)<sup>5</sup> due to the agreement between the experimentally determined  $g$  factor and the accepted literature value of  $g=2.0014$  for  $B_0$  parallel to the  $\langle 111 \rangle$  direction. In contrast to experiments at lower magnetic fields, the  $P_b$  resonance here is well separated from the two phosphorus resonances outside the field range that connects the two hyperfine peaks. This is expected as while the magnetic-field separation of  $P_b$  and phosphorus resonances depends on the  $g$ -factor difference, the phosphorus hyperfine splitting is constant ( $B=4.2$  mT) for high magnetic fields ( $B \gg 4.2$  mT).

### III. RESULTS OF EDMR MEASUREMENTS

The data show that the peak intensity  $A$  (defined as the integrated resonance lines=areas of the Gaussian fits) of the  $P_b$  resonance and the sum of the two hyperfine coupled  $^{31}\text{P}$  resonances have no correlation. While at  $T=12$  K, the  $P_b$  resonance is approximately equal to six times larger than the sum of the areas of the two  $^{31}\text{P}$  resonances, it is smaller at  $T=6$  K and  $T=3$  K. Moreover, at  $T=6$  K, the signs of the  $^{31}\text{P}$  resonances are positive, in contrast to the sign of the  $P_b$  resonance, which is consistently negative at all measured temperatures. These observations are in stark contrast to low-

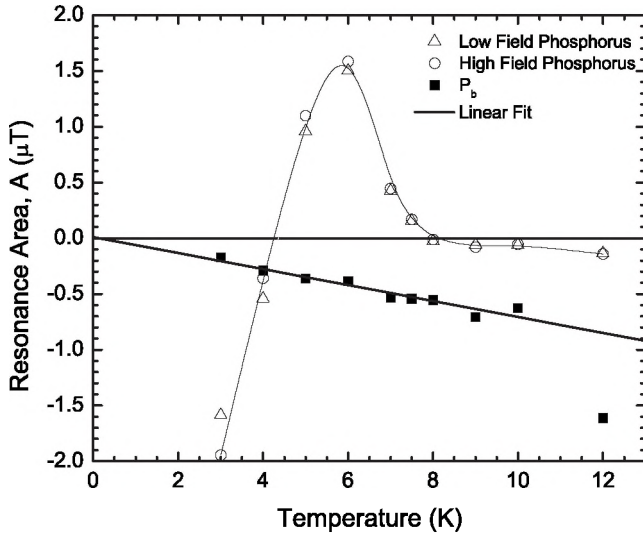


FIG. 3. Plot of the integrated area  $A$  under each of the three resonances as a function of the temperature. The linear fit to the  $P_b$  data between 3 and 10 K shows excellent agreement. The data at  $T=12$  K were taken at a different illumination intensity. The line joining the data for the  $^{31}\text{P}$  resonances is a guide to the eye.

field EDMR where the dominance of the  $^{31}\text{P}$ - $P_b$  pair mechanism causes a complete correlation between the intensities of  $P_b$  and the intensity sum of the two  $^{31}\text{P}$  peaks.<sup>7</sup> Additional evidence that different spin-dependent processes dominate at high magnetic fields is given by the current transient following a single short microwave pulse. Figure 2(b) shows such transients off resonance and on resonance for the phosphorus and the  $P_b$  peaks. These transients were taken with a pulse length of 8  $\mu\text{s}$ . A short microwave induced current which decays after approximately  $t=30$   $\mu\text{s}$  is seen in all traces. Additionally, a decrease in the current ( $\Delta I < 0$ ) is seen for both the phosphorus and  $P_b$  resonances. The return of the current to the steady-state value can in both cases be fit with a simple exponential decay. This is different from the more complex double exponential quenching/enhancement expected for the  $^{31}\text{P}$ - $P_b$  pairs that are visible at low magnetic fields. Additionally, the time constants of the two exponential recoveries [ $\tau_{P_b}=64(2)$   $\mu\text{s}$  and  $\tau_{\text{phos}}=23(2)$   $\mu\text{s}$  for  $P_b$  and phosphorus, respectively] are very different—further indicating that the two resonances are not due to the same processes. It shall be noted that the data in Fig. 2(b) show that EDMR at  $B_0 \approx 8.5$  T leads to a significantly smaller ratio between microwave induced artifact currents and spin-dependent currents than that seen at low fields. This makes high-field EDMR on silicon significantly more sensitive than X-band EDMR typically conducted at  $B_0 \approx 340$  mT.

#### IV. TEMPERATURE DEPENDENCE

In addition to the spectra displayed in Fig. 2, we have measured EDMR for a number of other temperatures between 3 and 10 K. Figure 3 shows the value of the peak areas of each of the three resonance peaks, namely,  $P_b$  and both the high- and low-field phosphorus. In order to obtain the maximum area, each peak was fit after correction of the lock-in

phase due to the different dynamic behaviors of the  $P_b$  and  $^{31}\text{P}$  signals. The magnitudes of the two phosphorus resonances are very similar as expected due to the negligible nuclear polarization. However, as foreshadowed in Fig. 2, the form of the temperature dependence is unexpectedly different from low-field EDMR experiments. From high to low temperature, the  $^{31}\text{P}$  signals are initially negative and smaller than the  $P_b$  signal, becoming larger and positive between  $T \approx 8$  K and  $T \approx 4.5$  K, before again becoming negative but with a larger magnitude for temperatures below  $T \approx 4.5$  K. We note that the sign and amplitude of the resonance at the lowest temperature recorded agrees with the single spectrum reported by Honig and Moroz.<sup>12</sup>

The experimental data presented allow us to exclude a number of mechanisms as the source for the observed signals and temperature dependencies. First, the signals observed are probably not bolometric effects due to resonant heating since this is expected to exhibit nonlinear monotonically decreasing temperature dependencies, which for both the  $^{31}\text{P}$  and the  $P_b$  signals are not in agreement with the observed conductivity changes. It is possible that the low-temperature ( $T < 6$  K)  $^{31}\text{P}$  signal has some bolometric components; however, Honig and Moroz<sup>12</sup> have assigned this to a spin-dependent neutral-donor capture and re-emission process based on a spectrum quantitatively similar to that presented here. Hence, we conclude that the signals observed must be due to spin-dependent electronic transport or recombination processes (except for  $^{31}\text{P}$  at temperatures  $T < 6$  K). Second, from the different magnitudes, the different signs, and the different temperature dependencies of the  $^{31}\text{P}$  and the  $P_b$  signals, we conclude that the  $^{31}\text{P}$ - $P_b$  interface recombination mechanism that dominates spin-dependent recombination rates at low magnetic fields is not responsible for the EDMR signals at high magnetic fields. Thus, the observed  $^{31}\text{P}$  and the  $P_b$  resonances must be due to independent electronic processes. The  $^{31}\text{P}$  enhancement signal for  $4.5 \text{ K} < T < 8$  K is not understood at this time in the absence of a theoretical framework describing this temperature behavior. This signal cannot be attributed to bolometric effects for the reasons stated above and because of its sign (as resonant sample heating is expected to cause a decrease in the conductivity). Thus, the source of the  $^{31}\text{P}$  enhancement signal cannot be attributed to an interface effect and consequently, the only signal that is clearly due to an interface process is the  $P_b$ -only transition.

#### V. MICROSCOPIC NATURE OF THE $P_b$ SIGNAL

We now consider the underlying process leading to the  $P_b$  resonance. As the  $P_b$  center is a paramagnetic deep interface state,<sup>5</sup> spin-dependent electronic transitions are described by a two spin-1/2 pair model.<sup>13,15</sup> Thus, the EDMR signal from the  $P_b$ -only transition can be due to: (i) Strongly coupled electron pairs such as the charged excited state  $P_b^{*}$  that decay spin-dependently into a charged ground state  $P_b^-$  (this model was first suggested by Friedrich *et al.*<sup>6</sup> for the low-field  $P_b$ -only signal observed at the interface of intrinsic  $c$ -Si to  $\text{SiO}_2$ ); (ii) tunneling; or (iii) energy-loss hopping between adjacent singly occupied  $P_b$  ground states with identical or

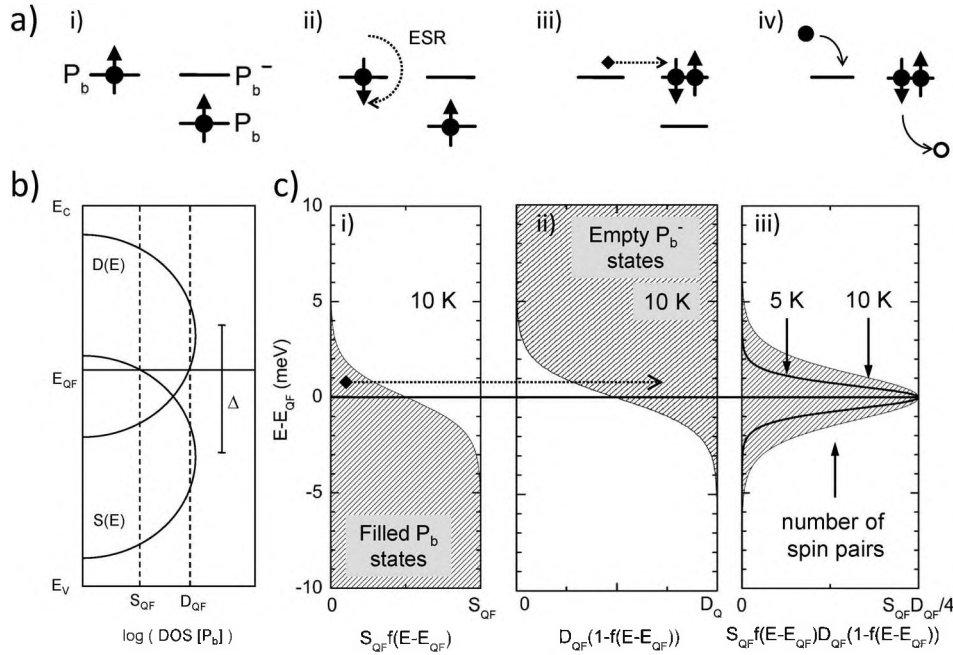


FIG. 4. (a) Cartoon of spin-dependent tunneling between two adjacent uncharged  $P_b$  centers where the ground-state energy of one  $P_b$  matches the  $P_b^-/P_b$  charging energy of the other. (i) Initially, the pair is in a triplet state and tunneling is not possible. (ii) Once the spin of one  $P_b$  state is changed the pair has singlet content and tunneling (iii) is allowed. (iv) An excess charge-carrier pair recombines as it discharges the two charged  $P_b$  states. (b) Sketch of DOS of  $P_b$  centers within the  $c$ -Si band gap (Ref. 5). Note that DOS's of  $P_b$  and  $P_b^-$  are assumed to be identical but shifted by the correlation energy  $\Delta$ . (c) Plot for  $T=10$  K of the number of (i) filled  $P_b$  states, (ii) filled  $P_b^-$  states, and (iii) tunneling pairs as a function of energy about the quasi-Fermi level  $E_{QF}$ . The sharper pair distribution for  $T=5$  K is indicated by the solid line. The plot illustrates that there are fewer pairs of adjacent  $P_b$  and  $P_b^-$  with matching energies when  $T$  decreases.

different energies, respectively. Figure 3 shows that the temperature dependence of the  $P_b$ -only signal can be well fit with a linear function without an offset (at  $T=0$  the EDMR signal  $\Delta I=0$ ) up to  $T \approx 10$  K. This behavior clearly contradicts model (i) as the EDMR signal is not expected to vanish for small  $T$  in this model. Models (ii) and (iii) involve transitions between adjacent  $P_b$  centers as illustrated for the tunneling case [model (ii)] in Fig. 4(a). Using Simmons-Taylor statistics<sup>19</sup> (an extension of Shockley-Read statistics to an arbitrary defect distribution of states) it can be shown that at finite temperature and under illumination, the occupancy of the  $P_b$  defects is given by the Fermi distribution  $f(E) = \{1 + \exp[-\frac{(E-E_{QF})}{k_B T}]\}^{-1}$ , about a quasi-Fermi energy  $E_{QF}$  with  $k_B$  the Boltzmann constant. The density of filled  $P_b$  states close to the quasi-Fermi energy is thus the  $P_b$  density of states (DOS),  $S(E)$ , times the Fermi distribution, i.e.,  $S(E)f(E)$ , as plotted in Fig. 4(c)(i) for  $T=10$  K. Similarly, the density of unfilled  $P_b^-$  states is given by  $D(E)[1-f(E)]$  and plotted in Fig. 4(c)(ii) for  $T=10$  K where  $D(E)$  is the  $P_b^-$  DOS. The  $P_b^-$  DOS is identical to the  $P_b$  DOS except that it is offset along the energy axis by the positive correlation energy  $\Delta$  associated with double occupancy of the defect<sup>5</sup> [i.e.,  $D(E) = S(E - \Delta)$ ] as is illustrated by the sketch of the DOS in the  $c$ -Si band gap shown in Fig. 4(b). We note that for the low-temperature range investigated here, the DOS is effectively constant [i.e.,  $S(E_{QF}) = S_{QF}$  and  $D(E_{QF}) = D_{QF}$ ] as the thermal energy is small ( $k_B T = 0.86$  meV at  $T = 10$  K) compared with the energy scale over which the DOS varies. Next, we assume that every  $P_b$  state interacts with  $P_b^-$  states within some

interaction radius  $r$ , which is independent of temperature. If we now consider only spin pairs whose energy levels are aligned [as expected for the tunneling model (ii)], we obtain a density  $n_p$  given by

$$n_p = \pi r^2 S_{QF} D_{QF} \int_{-\infty}^{\infty} f(E)[1-f(E)]dE = \xi T, \quad (1)$$

with the constant  $\xi = \pi r^2 S_{QF} D_{QF} k_B$ . From Eq. (1), we see that the density of spin pairs is linear in  $T$  with no pairs at  $T = 0$  K. As we anticipate a proportionality between the number of spin pairs and the EDMR signal, the energy-conserving model of tunneling between  $P_b$  pairs is in agreement with the observed temperature dependence of the EDMR signals. When we consider energy-loss hopping transitions [model (iii)], the spin-pair density becomes

$$n_p = \pi r^2 \int_{-\infty}^{\infty} S_{QF} f(E) \int_{-\infty}^E D_{QF} [1-f(E')]dE' dE \propto T^2, \quad (2)$$

in contrast to the experimental results.

We note that the transition from a  $^{31}\text{P}-P_b$  process<sup>7</sup> at low fields to a  $P_b$ -only mechanism at high fields may be explained by considering the underlying spin dynamics. The strength of an EDMR signal becomes weaker as the ratio  $\xi = \frac{\Delta\omega}{\gamma B_1}$  of the difference of the Larmor frequencies in a pair  $\Delta\omega$  to  $\gamma B_1$  drops below one with  $\gamma$  being the gyromagnetic ratio.<sup>20</sup> Hence, at low fields EDMR signals are dominated by maximized  $^{31}\text{P}-P_b$  signals as  $\xi > 1$  while  $\xi \ll 1$  for the  $P_b$  pairs with  $\Delta\omega \ll \gamma B_1$  due to the almost identical Landé fac-

tors of two  $P_b$  centers. At high fields, EDMR signals are dominated by  $P_b$  transitions since  $\xi > 1$  for both  $^{31}\text{P}-P_b$  and  $P_b-P_b$  pairs, while at the same time the  $P_b-P_b$  pair density is significantly higher than the  $^{31}\text{P}-P_b$  pair density.

## VI. SUMMARY

In summary, we have shown that EDMR on  $c\text{-Si:P}$  at the highest magnetic fields reported to date allows us to observe the influence of  $P_b$  centers and  $^{31}\text{P}$  donor atoms on spin-dependent photocurrents. In contrast to low magnetic-field EDMR, there is no intensity or transient correlation between these signals and, in contrast to the  $P_b$  signal, we find no evidence that the  $^{31}\text{P}$  signals are due to interface processes. The intensity of the  $P_b$  signal increases linearly with temperature, vanishing as  $T \rightarrow 0$ , which is shown to match the properties of charge-carrier tunneling between adjacent  $P_b$  states. This effect is expected only at high magnetic fields and does not contradict the dominance of the well investi-

gated  $^{31}\text{P}-P_b$  interface recombination process at low magnetic field. Finally, we point out that the spin-dependent tunneling demonstrated in this paper is analogous to the mechanism proposed by Kane<sup>3</sup> for readout of solid-state donor qubits. While previous attempts to investigate this mechanism have relied on remote charge detection of the transfer of an electron between clusters of donors<sup>21</sup> and even two single donors,<sup>22</sup> this work demonstrates a spin-dependent electronic tunneling transitions between localized defect sites. Note that the  $^{31}\text{P}-P_b$  mechanism that dominates at low magnetic fields does not demonstrate this effect as energy is not conserved.

## ACKNOWLEDGMENTS

This work was supported by Grant No. 7300-100 from the National High Magnetic Field Laboratory. G.W.M. was supported by the EPSRC through Grant Nos. GR/S23506 and EP/D049717/1.

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