# Two-spin relaxation of P-dimers in Silicon

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We study two-electron singlet-triplet relaxation of donor-bound electrons in Silicon. Hyperfine interaction of the electrons with the phosphorus (P) nuclei, in combination with the electron-phonon interaction, lead to relaxation of the triplet states. Within the Heitler-London and effective mass approximations, we calculate the triplet relaxation rates in the presence of an applied magnetic field. This relaxation mechanism affects the resonance peaks in current Electron Spin Resonance (ESR) experiments on P-dimers. Moreover, the estimated time scales for the spin decay put an upper bound on the gate pulses needed to perform fault-tolerant two-qubit operations in donor-spin-based quantum computers (QCs).

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Solid state based quantum information processing is a rapidly developing field, bridging quantum information science and condensed matter physics. Various schemes have been proposed to use spins as quantum bits (qubit), with the most prominent examples using electron spins in quantum dots[1] and $^{31}$P nuclear spins in Si.[2] Significant and exciting experimental progress have since been made to demonstrate coherent manipulation and measurement of spins in semiconductor nanostructures.[3–5]

It has been long demonstrated experimentally that single donor electron and nuclear spins in bulk Si have extremely long coherence and relaxation times.[6] Recent experiments on isolated donors show an electron spin decoherence time $T_2$ of 0.6 s at 1.8 K and 0.3 Tesla magnetic field (up to 10 s at press time).[7,8] This coherence time scale could be modified for donors near an interface, although measurements for single donors near an oxide interface have obtained a promising donor electron spin relaxation time of $T_1 \sim 6$ s at a magnetic field of 1.5 Tesla.[5]

The study of spin coherence properties of phosphorus dimers (two shallow P donors that are close to each other so that they are exchange coupled) in Si is relatively limited compared to those for single donors, because bulk experiments are ensemble averages over many different dimer configurations and couplings, while two-donor artificial devices are still in their infancy. P-dimers are an important ingredient in the spin-QC architectures that use the nuclear or electron spins of $^{31}$P in Si as qubits.[2,9] In these proposals exchange interaction between two bound electrons lead to an effective coupling between the corresponding electron or nuclear spins, which is used to implement two-qubit operations. Therefore, understanding spin coherence of P-dimers is crucial for the study of spin based quantum computing in Si:P.

Here we calculate singlet-triplet relaxation rates of donor-bound electrons in *bulk* silicon within the Heitler-London approximation.[10] In the absence of pure dephasing (such as that due to $^{29}$Si nuclear spins), such relaxation forms one of the decoherence channels for two-spin states of a dimer, and would give an upper bound to two-spin coherence. This relaxation process could also lead to broadening of resonance peaks in an ESR measurement of P-dimers, making it an observable effect within the currently available spin resonance technology.

*Theoretical Model.* We consider two electrons which are bound to two phosphorus donors (P dimer), separated by a distance $2d$, as shown in Fig. 1a. The crystallographic axes of silicon are denoted by $x$, $y$ and $z$. The direction of the applied magnetic field is along $Z$ (not necessarily $z$) axis, which defines the quantization axis in the spin space. The Hamiltonian of the coupled electrons-nuclei system is then given by $H = H_0 + V_{HF}$ where

$$
\begin{aligned}
H_0 &= g\mu_B B(\sigma_Z^{1e} + \sigma_Z^{2e}) - g_n\mu_n B(\sigma_Z^{1n} + \sigma_Z^{2n}) \\
&\quad + J\boldsymbol{\sigma}^{1e} \cdot \boldsymbol{\sigma}^{2e} , \tag{1}
\end{aligned}
$$
$$
V_{HF} = A_{HF} \sum_{i,j=1,2} \boldsymbol{\sigma}^{in} \cdot \boldsymbol{\sigma}^{je} \delta(\boldsymbol{r}_j - \boldsymbol{R}_i), \tag{2}
$$

$g$ ($g_n$) is the effective g-factor of the electron (nucleus), $\mu_B$ ($\mu_n$) is the electron (nuclear) magnetic moment, $J$ denotes the Coulomb exchange interaction between the two electrons, and $A_{HF} = 0.2$ $\mu$eV is the hyperfine coupling constant between a phosphorus nucleus and its bound electron.[2] The superscripts $e$ ($n$) refers to the electron (P nucleus), and 1 and 2 label the first and the second electron (nucleus). Notice that the total electron spin is not a good quantum number due to the hyperfine interaction $V_{HF}$. For example, if the two nuclear spins are anti-aligned, the two electrons would experience an inhomogeneous magnetic field, which mixes the electron singlet and triplet states.[11] This mixing then allows a phonon-mediated relaxation between the two-electron spin states, and is at the core of the present study.

Using the Heitler-London approximation and averaging over the electron orbitals, we simplify the above Hamiltonian by introducing the averaged hyperfine coupling $A$: $\langle\psi_{as}|V_{HF}|\psi_{as}\rangle \simeq \langle\psi_s|V_{HF}|\psi_s\rangle \simeq \frac{1}{2}A(\boldsymbol{\sigma}^{1n} + \boldsymbol{\sigma}^{2n}) \cdot (\boldsymbol{\sigma}^{1e} + \boldsymbol{\sigma}^{2e})$ and $\langle\psi_{as}|V_{HF}|\psi_s\rangle = \langle\psi_s|V_{HF}|\psi_{as}\rangle \simeq \frac{1}{2}A(\boldsymbol{\sigma}^{1n} - \boldsymbol{\sigma}^{2n}) \cdot (\boldsymbol{\sigma}^{1e} - \boldsymbol{\sigma}^{2e})$, where the symmetric and antisymmetric electronic orbital wave functions are denoted by $\psi_s$ and $\psi_{as}$, respectively. Considering only the ground orbital state on each donor, $\psi_{a,as} = \{\psi_L(\boldsymbol{r}_1)\psi_R(\boldsymbol{r}_2) \pm$

$\psi_L(\boldsymbol{r}_2)\psi_R(\boldsymbol{r}_1)\}/\sqrt{2}$, where $\psi_L$ ($\psi_R$) is the ground orbital wave function of the left (right) donor electron to be specified later. We note that the first excited state of the donor bound electron is about 10 meV above the ground state[12], and we do not consider dimers that are too tightly coupled ($d \geq 8$ nm), so that the Heitler-London approximation is justified for our calculation.

Within the above mentioned approximations, the $16 \times 16$ spin Hamiltonian of the two electrons and two nuclei reduces to six clusters (one quartet, five doublets, and two singlets) that are block-diagonal. The effective Hamiltonian for the quartet cluster of the electrons Hilbert space, in $\{S^e S^n,\ T_0^e T_0^n,\ T_+^e T_-^n,\ T_-^e T_+^n\}$ bases, reads

$$H_{eff} = \begin{pmatrix} -3J & 2A & -2A & -2A \\ 2A & J & 2A & 2A \\ -2A & 2A & J_1 & 0 \\ -2A & 2A & 0 & J_2 \end{pmatrix}, \quad (3)$$

$$J_1 = J + 2g\mu_B B + 2g_n\mu_n B - 2A, \quad (4)$$
$$J_2 = J - 2g\mu_B B - 2g_n\mu_n B - 2A, \quad (5)$$

where $\{S, T_0\} = \frac{1}{\sqrt{2}}|\uparrow\downarrow\rangle \mp |\downarrow\uparrow\rangle$ and $\{T_+, T_-\} = \{|\uparrow\uparrow\rangle, |\downarrow\downarrow\rangle\}$ are the singlet and the triplet spin states of the electrons and the phosphorus nuclei, as shown in Fig. 1b. The effective Hamiltonians for the remaining doublets are given by

$$H^{(2)} = \begin{pmatrix} -3J & 2A \\ 2A & J \end{pmatrix}, \{S^e T_0^n, T_0^e S^n\},$$

$$H^{(3)} = \begin{pmatrix} -3J + 2g_n\mu_n B & 2A \\ 2A & J - 2g\mu_B B \end{pmatrix}, \{S^e T_-^n, T_-^e S^n\},$$

$$H^{(4)} = \begin{pmatrix} -3J - 2g_n\mu_n B & 2A \\ 2A & J + 2g\mu_B B \end{pmatrix}, \{S^e T_+^n, T_+^e S^n\},$$

$$H^{(5)} = \begin{pmatrix} J + 2g_n\mu_n B & 2A \\ 2A & J - 2g\mu_B B \end{pmatrix}, \{T_0^e T_-^n, T_-^e T_0^n\},$$

$$H^{(6)} = \begin{pmatrix} J - 2g_n\mu_n B & 2A \\ 2A & J + 2g\mu_B B \end{pmatrix}, \{T_0^e T_+^n, T_+^e T_0^n\}.$$

The fully polarized states, $\{T_+^e T_+^n\}$ and $\{T_-^e T_-^n\}$, are decoupled from the remaining states.

In the following we neglect the Zeeman splitting of the phosphorus nuclear spins because it is about three orders of magnitude smaller than the electron Zeeman splitting. We also assume that the averaged hyperfine coupling is much smaller than the exchange energy $A \ll J$ to keep our perturbation treatment valid, so that level alignment is qualitatively given by Fig. 1.b.

*The electron-phonon interaction.* Direct relaxation of electron spin states are generally forbidden since electron-phonon interaction conserves spin. However, as shown in Eq. (3), electron spin singlet and triplets are mixed via the hyperfine interaction in a P-dimer. At low energies ($J \leq 1$ meV), which is our main concern here, only acoustic phonons contribute to the spin relaxation processes. Moreover, only *deformation potential* coupling, consisting of a *dilation* part and a *shear* part, contribute to the
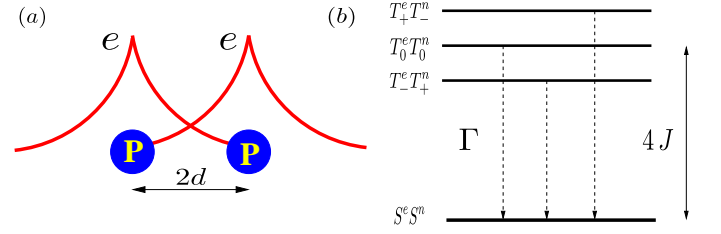


FIG. 1: (a) Schematic of a P-dimer and the corresponding (hydrogenic) electrons envelope functions. (b) Energy levels of the two electrons bound to a P-dimer in the quartet cluster, see Eq. (3). Mixing of the donor electron eigenstates is achieved via the hyperfine coupling to the P nuclear spins, which causes the triplet-singlet relaxation of the electron spins via emission of acoustic phonons. Here $\Gamma$ refers to different relaxation rates from triplet states and $4J$ is the splitting between $T_0^e$ and $S^e$.

relaxation processes in Si (or Ge). The Hamiltonian for an electron in one of the valley minima of Si is[13,14]

$$H_{HV}^D = \Xi_d \operatorname{Tr}\{e\} + \Xi_u(\hat{\boldsymbol{k}} \cdot \boldsymbol{e} \cdot \hat{\boldsymbol{k}}), \quad (6)$$

where $\boldsymbol{e}$ is the strain tensor due to phonons and $\hat{\boldsymbol{k}}$ is the unit vector along the direction of one of the [100] conduction band minima in the reciprocal space. $\Xi_u$ is the shear deformation potential due to uniaxial strain along the [100] direction and $\Xi_d + \Xi_u$ is the volume deformation potential[14]. The electron-phonon interaction for a donor electron in the ground state is then given by[15]

$$H_{ep}^D = (\Xi_d + \frac{1}{3}\Xi_u) \sum_{\boldsymbol{q}} \frac{q\, e^{i\boldsymbol{q}\cdot\boldsymbol{r}}}{\sqrt{2\rho\omega_{\boldsymbol{q}}/\hbar}} (a_{\boldsymbol{q}} + a_{-\boldsymbol{q}}^\dagger), \quad (7)$$

where $a_{\boldsymbol{q}}$ annihilates a phonon with momentum $\boldsymbol{q}$, $\rho = 2330$ kg/m$^3$ is the silicon density, $\omega_{\boldsymbol{q}} = v_l q$ is the energy of the longitudinal acoustic phonon, $v_l$ is its corresponding velocity and we only consider the longitudinal phonons which have the dominant contribution to the electron-phonon interaction. We note that in Si, $\Xi_d \sim 5.0$ eV and $\Xi_u = 8.77$ eV.[14]

*The relaxation rate.* To calculate two-spin relaxation, we need the two-electron orbital wave functions for the P-dimer. The conduction band of Si has six degenerate minima located close to (but not at) the edge of the first Brillouin zone.[14] In the effective mass approximation, the ground state wave function of the donor bound electron is given by the homogeneous superposition of these six valleys

$$\Psi(\boldsymbol{r}) = \frac{1}{\sqrt{6}} \sum_{\mu=1}^{6} F_\mu(\boldsymbol{r})\phi_\mu(\boldsymbol{r}), \quad (8)$$

$$\phi_\mu(\boldsymbol{r}) = u_\mu(\boldsymbol{r})e^{i\boldsymbol{k}_\mu\cdot\boldsymbol{r}}, \quad (9)$$

where $\mu = \{\pm x, \pm y, \pm z\}$ is the valley index, $\phi_\mu(\boldsymbol{r})$ are the Bloch wave functions and $F_\mu(\boldsymbol{r})$ are their corresponding

TABLE I: Relaxation time $T_1$ (ms) for $T_0^e T_0^n \rightarrow S^e S^n$ (see Fig. 1.b) as a function of $J$ (meV) and $d$ (Å) for *substitutional* donors located along [001], [011] and [111] axes. $a_0 = 5.43$ Å is the lattice constant in Si, $a_1 = a_0/\sqrt{2}$ and $a_2 = \sqrt{3}a_0/4$.

| $2d/a_0$ | 15 | 16 | 17 | 18 | 19 | 20 | 21 | 22 |
|---|---|---|---|---|---|---|---|---|
| $4J$ | 1.19 | 0.76 | 0.57 | 0.47 | 0.38 | 0.29 | 0.2 | 0.13 |
| $T_1$ | 0.02 | 0.04 | 0.08 | 0.18 | 0.41 | 1.0 | 2.6 | 7.5 |
| $2d/a_1$ | 23 | 24 | 25 | 26 | 27 | 28 | 29 | 30 |
| $4J$ | 0.25 | 0.26 | 0.03 | 0.3 | 0.01 | 0.15 | 0.03 | 0.04 |
| $T_1$ | 0.15 | 0.2 | 2.5 | 0.35 | 17.5 | 2 | 13 | 315 |
| $2d/a_2$ | 36 | 37 | 40 | 41 | 44 | 45 | 48 | 49 |
| $4J$ | 0.26 | 0.25 | 0.34 | 0.015 | 0.06 | 0.11 | 0.006 | 0.04 |
| $T_1$ | 0.08 | 0.11 | 0.18 | 5 | 2.9 | 2 | 72 | 13.5 |

envelope functions. To find a simple analytical expression for the spin decay rate, we first assume Gaussian envelope functions for the donor electrons

$$F_{\pm z}^G(\boldsymbol{r}) = \left(\frac{2}{\pi}\right)^{3/4} \frac{1}{\sqrt{a^2 b}} \, e^{-\frac{x^2+y^2}{a^2} - \frac{z^2}{b^2}}, \quad (10)$$

where $a$ and $b$ are the effective Bohr radii, and the remaining envelope functions are obtained by cyclic change of $x$, $y$ and $z$ in Eq. (10). Using the Fermi golden rule and Eq. (7), we calculate the relaxation rate (the one-phonon process) from $T_0^e$ to $S^e$ and obtain

$$\Gamma_{T_0 \rightarrow S} \approx \alpha_1 (\Xi_d + \tfrac{1}{3}\Xi_u)^2 \, A^2 J (n_{q_0}+1)|I|^4, \quad (11)$$

$$\Gamma_{T_0 \rightarrow S}^G \approx \alpha_2 (\Xi_d + \tfrac{1}{3}\Xi_u)^2 \, A^2 J (n_{q_0}+1) \times \\ \left[ e^{-d^2/b^2} + 2e^{-d^2/b^2} \right]^4, \quad (12)$$

where $\alpha_1 \simeq 100\,\alpha_2 = 25/\pi^2 \rho \hbar^4 v_l^5$, $q_0 = 4J/\hbar v_l$, $I = \langle \Psi_L(\boldsymbol{r})|\Psi_R(\boldsymbol{r})\rangle$ is the overlap integral of the right and left donor wave functions, $n_q$ is the Bose-Einstein distribution of phonons, and $\Gamma^G$ is the relaxation rate for a Gaussian envelope function. The relaxation rates $\Gamma_{T_+ \rightarrow S}$ and $\Gamma_{T_- \rightarrow S}$ have the same form as in Eq. (11) but with the following replacement $J \rightarrow (J_{1,2} + 3J)/4$ [see Eqs. (4,5)]. Clearly, $\Gamma$ has an explicit linear dependence on the exchange energy $J$ and a quadratic dependence on the hyperfine coupling $A$.

Since in general $J \propto I^2$, one might conclude that $\Gamma$ should scale as $J^3$. However, the exchange integrals are non-trivial functions of the distance between the donor sites $d$ and have oscillatory dependence on donor positions,[16] so that a simple polynomial scaling is generally not available.

A more realistic envelope function $F_\mu(\boldsymbol{r})$ for a donor should be hydrogenic with anisotropy. For example, Kohn and Luttinger proposed a variational form[12]

$$F_{\pm z}(\boldsymbol{r}) = \frac{1}{\sqrt{\pi a^2 b}} \, e^{-\sqrt{\frac{x^2+y^2}{a^2} + \frac{z^2}{b^2}}}. \quad (13)$$
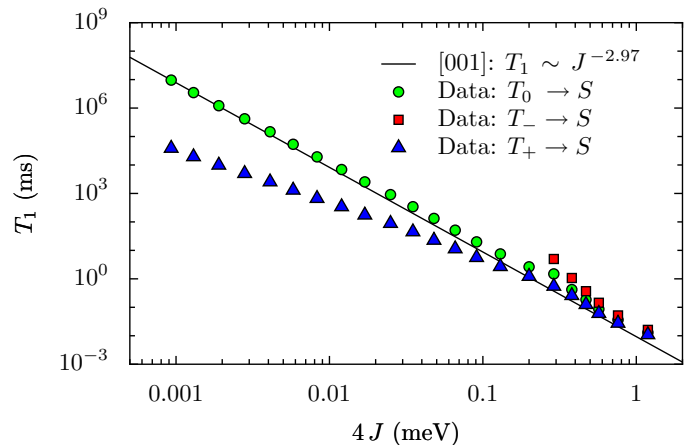


FIG. 2: (Color online) *Almost*-cubic dependence of the relaxation rate $\Gamma$ of the symmetric two-electron triplet state $T_0^e$ on the singlet-triplet splitting $4J$, for substitutional donors along [001] direction, see Eq. (11). An external magnetic field $B = 1$ T is applied, in order to split the triplet states. Note that for $T_-^e$ state, we took only those $J$s which were larger than the Zeeman energy.

With this envelope function there is no closed analytical form for the overlap integrals and the electron-phonon matrix elements. We therefore calculate the relaxation rates numerically. Table I shows our results for the relaxation rate of the unpolarized triplet to singlet state, where we have taken into account the oscillatory behavior of $J$ as a function of the donors separation $2d$. The relaxation times change from tens of $\mu$s to hundreds of ms as we vary the distance between the substitutional donors $2d$ (and their relative orientation) from 8 to 12 nm. We also note that the value of the exchange, as a function of the donor sites, strongly depends on along which symmetry axes of the crystal the two donors are aligned.[16] We find an almost cubic dependence of the relaxation rate on $4J$ for donors along [001] direction, as shown in Fig. 2. On the other hand, there is no reliable polynomial fit for the other two directions [011] and [111].

Our calculation for $J$ is based on the *plane wave approximation*[17] of the Bloch wave function $\phi(\boldsymbol{r}) = u_{\boldsymbol{k}}(\boldsymbol{r})e^{i\boldsymbol{k}\cdot\boldsymbol{r}}$, where we only keep the plane wave part and neglect the lattice periodic part $u_{\boldsymbol{k}}(\boldsymbol{r})$. The numerical values of $J$ do not differ significantly (specifically in the Heitler-London approximation) from the more elaborate approaches which incorporate the full Bloch structure of the Kohn-Luttinger electron wave functions[17,18]. Therefore, our final results for the relaxation times $T_1$ give the right order of magnitude due to the linear dependence of the rates on $J$. We stress that our calculations are valid in the regime of relatively large exchange energies $A \ll J$. In other words, although $J$ is oscillatory as a function of $d$, it remains always finite (over a large range of values) and larger than the hyperfine coupling in [100]
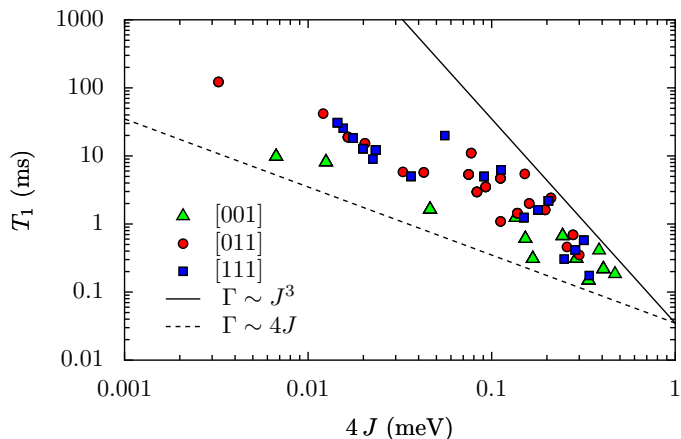
FIG. 3: (Color online) Two-spin $T_0 \to S$ relaxation times as a function of the exchange splitting for donor pairs, which are located about 10 nm apart from each other and along different symmetry axis. The position of one of the donors varies within its nearest neighboring sites, in a radius 5 Angstroms. The solid and dotted lines show that the resulting data is within two polynomial curves of $J^3$ and $J$.

direction. In contrast, for donors located apart along [110] or [111] axes, there are certain values of $d$ which result in (almost) zero exchange energy[16] where our perturbation scheme breaks down.

In reality, it is challenging to implant donors at specific lattice sites. Figure 3 shows the two-spin relaxation time, calculated with Eq. (11), as a function of exchange splitting $J$, for donor pairs which are about 10 nm apart (along different symmetry axis), with the position of one of the donors varying within a radius of 5 Angstrom. Our results show that even with this small fluctuation in the donor location, there could be a three-orders-of-magnitude variation in both exchange splitting and triplet-singlet relaxation rate, ranging from 0.1 ms to 100 ms.

The main result of our calculations is that even in purified samples (with no [29]silicon isotopes), the two-electron states can relax via the combined effect of the hyperfine interaction between phosphorus nuclei and the electrons, and the electron-phonon interaction.[19] Furthermore, this relaxation mechanism is strongly dependent on the donor separation and their orientation along different symmetry axes of the crystal. Our results provide an upper limit to the duration of the two-qubit gates in the Kane architecture,[2] where the two phosphorus nuclei are entangled indirectly via the bound electrons through the hyperfine and exchange interaction. Due to the strong dependence of the relaxation time of the electronic triplet states on the location (and orientation) of the donors,

the physical realization of donor-based schemes requires a careful control over the donor positions.

The data presented in Table I and Figs. (2,3), as well as the expressions for the relaxation rate $\Gamma$, allow the identification of optimal separations and orientations for substitutional donors which mitigate this relaxation mechanism, yet still permit sufficiently fast two-qubit operations. The key is that the relaxation rate is proportional to $J|I|^4$, while the speed of two-qubit gates depends on $J$ linearly, whether for electron or nuclear spin qubits. The additional dependence on inter-donor overlap means that at larger inter-donor separations, or for those pair-positions that have reduced overlap due to valley interference, a larger ratio of $J/\Gamma$ should be available, so that fault-tolerant two-qubit gates are possible. For nuclear spin qubits more specifically, we note that the use of the electron spins as intermediaries for two-qubit gates[2] could have significant impact on gate fidelity. While isolated P nuclear spins are known to have outstanding coherence properties,[6] the electron spin relaxation mechanism studied here could put a much stronger constraint on the gate speed, making the electron spin coherence properties the ultimate gauge to determine the feasibility of nuclear spin qubits for quantum information processing.

In the electron spin spectroscopy for P dimers, transitions between the electron spin triplet states are detected, while singlet states are not directly involved. The finite relaxation calculated here from triplet states to the singlet state presents a leakage for the triplet populations, thus should lead to a broadening of the ESR signal for the dimers.

We note that the effects of the spin-orbit interaction can be safely neglected due to the small spin-orbit coupling in silicon.[20] However, for shallow donors located close to an interface, the interface roughness could potentially lead to a sufficiently strong electric field near the bound electrons, which would in turn induce a non-negligible extrinsic spin-orbit interaction. Due to the lack of a comprehensive model of the interface and its effect on the spin of the electrons, we did not cover this issue in this work and instead focused on bulk properties.

In conclusion, we have calculated the relaxation rates of the electronic triplet states to the singlet state in P-dimers. These rates have nontrivial dependence on the singlet-triplet exchange splitting, and are strongly anisotropic. This relaxation mechanism can be studied in P-dimers spin spectroscopy experiments by measuring the linewidth of the ESR peaks. Finally, our results can be easily extended to similar materials (like Ge) with different valley degeneracies.

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