

Donor-Driven Spin Relaxation in Multivalley Semiconductors

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The observed dependence of spin relaxation on the identity of the donor atom in *n*-type silicon has remained without explanation for decades and poses a long-standing open question with important consequences for modern spintronics. Taking into account the multivalley nature of the conduction band in silicon and germanium, we show that the spin-flip amplitude is dominated by short-range scattering off the central-cell potential of impurities after which the electron is transferred to a valley on a different axis in *k* space. Through symmetry arguments, we show that this spin-flip process can strongly affect the spin relaxation in all multivalley materials in which time-reversal cannot connect distinct valleys. From the physical insights gained from the theory, we provide guidelines to significantly enhance the spin lifetime in semiconductor spintronics devices.

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Electrical spin injection from ferromagnetic metals to silicon (Si) is imperative for the operation of spintronics devices that can be integrated with mainstream semiconductor technology [1–3]. Because of the so-called conductivity and spin lifetime mismatch problem, one cannot use Ohmic contacts between semiconductors and metals for electrical injection of spin-polarized currents [4–6]. Accordingly, electrical techniques are largely limited to spin injection by ballistic hot electrons [7–11], or tunneling across thin barriers [12–16]. The latter approach can be integrated with mainstream fabrication techniques due to the built-in potential barrier formed by the depletion region in the semiconductor side of direct metal-semiconductor Schottky contacts or of metal-oxide-semiconductor junctions with ultrathin oxide layers. Effective tunneling requires interface doping with donor concentrations of 10^{19} cm⁻³ or higher so that the built-in potential barrier is only a few nanometers wide [3,12,13]. These degenerate doping levels come with a penalty of enhanced spin relaxation due to electron-impurity scattering. To date, there is no theory that can quantify the spin relaxation in degenerate *n*-type silicon [1,17], or explain its empirically found strong dependence on the donor identity [18–22]. This void enabled recent spin injection experiments to claim significantly shorter spin lifetimes than those of well-established electron paramagnetic resonance (EPR) experiments [3,23]. Clearly, a theory that identifies and quantifies the dominant spin relaxation process can resolve the apparent contradiction.

In this Letter, we address these long-standing open questions finding that the dominant spin-flip process takes place when conduction electrons are scattered off the donor core potential, after which they are transferred to a valley on a different axis in *k* space. This process is clarified by using the scattering symmetry in the presence of spin-orbit coupling, an approach that has been overlooked in

multivalley crystals ever since the early studies of impurities' core-potential effects on mobility in semiconductors [24] or on spin flips in metals (e.g., Ref. [25]). We elucidate the general spin-flip process for Si where available experimental data can be used to test its validity, and explicitly extend the analysis to the case of germanium. Through symmetry arguments, we discuss ramifications of this spin-flip process finding it can strongly affect the spin relaxation in all multivalley materials where time reversal cannot connect distinct valleys. Unveiling this long-awaited explanation allows us to predict a method to prolong the spin lifetime by lifting the valley degeneracy, thereby enabling important progress in spintronics.

A salient feature of spin relaxation in *n*-type silicon is a strong dependence on the donor atom identity [18–22]. It has been long recognized from electron paramagnetic resonance experiments that the spin lifetime is about 100 times shorter in heavily antimony-doped silicon (Si:Sb) than in phosphorus-doped silicon (Si:P) with comparable impurity concentration [20,22]. This finding contradicts the traditional Elliott picture for spin relaxation, in which the probability for an electron to flip its spin is governed by the spin-orbit coupling of the host material (Si in this example), whereas the identity of the scattering center is of little importance [26–28]. In addition, the predicted proportionality between charge mobility and spin relaxation time seems at odds with empirical values in *n*-type Si. That is, the spin relaxation is markedly different in Si:P, Si:As, or Si:Sb with comparable impurity concentration [18–22], while the mobility is essentially the same [29–31].

The short-range potential of impurities, rather than the spin mixing of states in the host materials, governs the strong dopant-dependent spin relaxation. In particular, we will show that this trend is dominated by the difference between the potentials of the impurity and host atoms in the

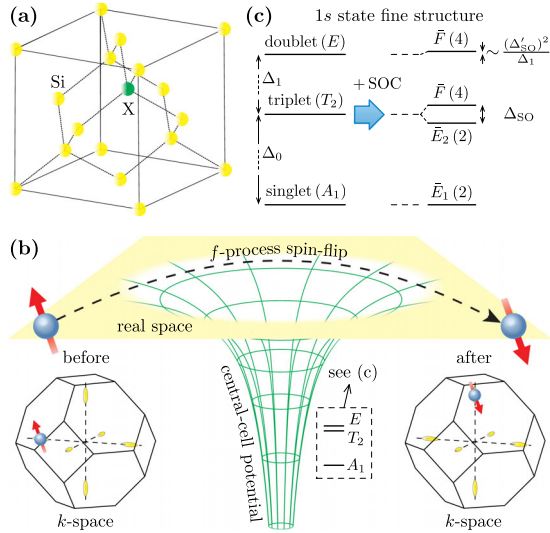


FIG. 1 (color online). (a) Substitutional impurity atom in a Si crystal host. We consider typically employed group V donors such as $X = \{P, As, Sb\}$. (b) Scheme of the dominant impurity-driven spin relaxation mechanism. The spin flip is governed by scattering off the central-cell potential after which the conduction electron is transferred to a valley on a different crystal axis in k space. (c) Fine structure of the $1s$ state due to the central-cell potential. The scattering amplitude in (b) is governed by the impurity fine-structure parameters Δ_{so} and Δ'_{so} .

central-cell region, due to spin-orbit coupling (SOC) within the immediate vicinity of the impurity atomic core. Figure 1(a) shows an example for a substitutional impurity atom surrounded by four host atoms in a tetrahedral molecular geometry. The vast majority of donors and acceptors in Si are represented by such substitutional impurities whose potential has T_d point-group symmetry. From this symmetry and the multivalley nature of the Si conduction band, we obtain from a rigorous zeroth-order wave vector analysis of group theory that the f -process intervalley scattering is the only nonvanishing spin-flip scattering mechanism (i.e., between two valleys residing on different crystallographic axes [32]). This process is schematically shown in Fig. 1(b). The impurity-induced spin relaxation is evidently weaker for intravalley scattering in which the electron remains in the same valley, or for the intervalley g process in which the electron is transferred to the opposite valley on the same crystallographic axis. Their vanishing amplitudes can be shown by invoking C_2 -rotation operations of the T_d group or time reversal symmetry, respectively (i.e., after such symmetry operations the scattering matrix element is negated).

Another aspect of the theory relies on the fundamental relation between the scattered and bound states of an impurity potential [33]. For the small region around the ion core, the impurity $1s$ wave functions are basically the same as the corresponding conduction valley edge wave functions, differing only by the volume normalization due to the hydrogenic envelope of the former. This relation allows us to quantify the spin-flip amplitude from the

empirically known SOC-induced splitting of the donor-state spectral lines. The left part of Fig. 1(c) shows the energy levels of donor states in Si. Because of the valley-orbit coupling within the T_d impurity, this $1s$ state is split into spin-independent nondegenerate (A_1), doubly degenerate (E), and triply degenerate (T_2) states where the overall sixfold multiplicity comes from the number of conduction edge states (valley centers) [34]. A_1 , E , and T_2 denote the symmetrized linear combinations of these valley edge states under T_d group operations [e.g., $\psi_{A_1} = (1, 1, 1, 1, 1, 1)/\sqrt{6}$ where each vector element corresponds to one valley edge]. Taking into account the SOC of the impurity, two important energy scales are relevant for spin relaxation. The twelve spin-dependent states are symmetrized in T_d group

$$A_1 \times \bar{E}_1 = \bar{E}_1, \quad E \times \bar{E}_1 = \bar{F}, \quad T_2 \times \bar{E}_1 = \bar{F} + \bar{E}_2, \quad (1)$$

where a pure spin transforms as \bar{E}_1 representation. The first spin relaxation constant corresponds to splitting of the T_2 state to fourfold (\bar{F}) and twofold (\bar{E}_2) spin dependent states, as shown in the right part of Fig. 1(c). The SOC-induced splitting is known empirically: $\Delta_{so} \approx 0.03$ meV for Si:P, 0.1 meV for Si:As, and 0.3 meV for Si:Sb [35,36]. The second energy scale is more subtle and comes from spin-dependent interaction between the fourfold degenerate \bar{F} states, stemming from E and T_2 spinless states, respectively [two \bar{F} levels in Fig. 1(c)]. As derived in the Supplemental Material [37], this interaction is manifested by a small added contribution, Δ'_{so} , to the splitting of these states

$$\varepsilon_{\bar{F}} \rightarrow \frac{1}{2} \left[(\varepsilon_{T_2} + \varepsilon_E) \pm \sqrt{\Delta_1^2 + (\Delta'_{so})^2} \right]. \quad (2)$$

$\varepsilon_{T_2(E)}$ is the spin-independent energy of the T_2 (E) state where $\Delta_1 = \varepsilon_{T_2} - \varepsilon_E$ is their valley-orbit induced splitting (typically much larger than Δ'_{so}). The presence of Δ'_{so} is mandated by symmetry as two \bar{F} states are inevitably coupled by T_d symmetric scatterers and its magnitude should be comparable with that of Δ_{so} since both originate from similar impurity orbitals (the two sets of specific \bar{F} state vectors for the symmetry-allowed coupling are explicitly shown in Ref. [37]). Indeed, we will show that the theory agrees with empirical values of the spin relaxation for $|\eta| \equiv |\Delta'_{so}/\Delta_{so}| \approx 2$.

Using Δ_{so} and η , we quantify the spin relaxation in heavily doped n -type Si due to scattering off the impurity central-cell potential. Derivation details are found in the Supplemental Material [37] and here we summarize the main findings. The scattering matrix elements are derived for transitions between opposite-spin states of the conduction band minima (zeroth-order wave vector analysis). By changing the symmetry-adapted T_d state basis into the basis of individual valley states [e.g., $\psi_{+x} = (1, 0, 0, 0, 0, 0)$ denotes the edge state of the $+x$ valley], we convert the symmetrized scattering process into intravalley, g and f processes. We find only the latter has a nonvanishing amplitude [an example of this f process is

schematically shown in Fig. 1(b)]. The resulting dominant spin-flip scattering amplitude reads [37]

$$U_{sf}^f = \frac{\pi a_B^3}{V} \left[\frac{i e^{i\phi}}{6} \sin\theta + \frac{\eta(1+i)}{\sqrt{12}} \left(\cos^2 \frac{\theta}{2} - i \sin^2 \frac{\theta}{2} e^{2i\phi} \right) \right] \Delta_{so}, \quad (3)$$

where V is the crystal volume and $a_B \approx 2$ nm is the electron Bohr radius in Si. The normalization factor $\pi a_B^3/V$ is due to the fact that Δ_{so} and η are bound-state parameters whereas the transition amplitude is that of scattered states normalized over the crystal. The polar and azimuthal angles (θ & ϕ) define the spin orientation, where the polar angle is measured from the normal direction to the plane defined by two valley axes of the f process. To better understand this angular dependence, we consider an example where electrons have a net spin polarization along the z axis. The spin-flip amplitude is calculated by assigning $\theta = 0$ for scattering between $\pm x$ and $\pm y$ valleys since the net spin polarization along the z axis is parallel to the normal of the xy plane. Similarly, we assign $\theta = \pi/2$ & $\phi = 0$ ($\theta = \pi/2$ & $\phi = \pi/2$) for scattering between $\pm z$ and $\pm x(y)$ valleys since the net spin polarization along the z axis is perpendicular to the normal of the xz (yz) plane. After averaging over all valley configurations and summing over final states, we get that the spin relaxation of a conduction electron with energy $\epsilon_{\mathbf{k}}$ above the band edge is

$$\begin{aligned} \frac{1}{\tau_s(\epsilon_{\mathbf{k}})} &= \frac{4\pi N_d V^2}{\hbar (2\pi)^3} \int d^3 k' |U_{\mathbf{k}\uparrow\mathbf{k}'\downarrow}(\mathbf{s})|^2 \delta(E_{\mathbf{k}'} - \epsilon_{\mathbf{k}}) \\ &= \frac{4\pi N_d m_e a_B^6}{27\hbar^4} \sqrt{2m_e \epsilon_{\mathbf{k}}} (6|\eta|^2 + 1) \Delta_{so}^2. \end{aligned} \quad (4)$$

where $U_{\mathbf{k}\uparrow\mathbf{k}'\downarrow} = U_{sf}^f$ in Eq. (3), N_d is the donor concentration, and $m_e = 0.32m_0$ is the electron effective mass in Si. In the high-temperature regime, we can assign $\epsilon_{\mathbf{k}} \approx k_B T$ and get that the effective spin relaxation rate scales with \sqrt{T} . In the opposite limit ($\epsilon_F \gtrsim k_B T$), the average spin lifetime is temperature independent and found by assigning $\epsilon_{\mathbf{k}} \approx \epsilon_F$ where $\sqrt{2m_e \epsilon_F}/\hbar \approx (3\pi^2 N_d)^{1/3}$. The solid lines in Fig. 2(a) show the calculated spin lifetime in this limit for $|\eta| = 2$. The symbols are compiled results from seven different experiments [15,18–22,39]. The theory shows excellent agreement with experiment apart from a small discrepancy for Si:P when approaching the critical metal-to-insulator transition ($\sim 0.2\text{--}0.4 \times 10^{19} \text{ cm}^{-3}$). The relatively long spin lifetime of Si:P in this regime may have another contribution due to remnant effects of the impurity band [40]. For the case of n -type Si:Sb, we could not find experimental results for $N_d > 10^{19} \text{ cm}^{-3}$ possibly due to strong antimony segregation when growing samples at these doping levels [41].

In contrast to the spin relaxation of conduction electrons, their mobility is not affected by the identity of the donor atom. Figure 2(b) shows a compilation of empirical mobility values along with the theoretical curve [30]. The mobility is governed mostly by the potential tail of

ionized impurities away from the central cell, $U(r) \propto \exp(-\kappa r)/r$ where κ^{-1} is the screening length. This potential tail is identical for donors from the same column of the periodic table, explaining why the proportionality factor between mobility and spin relaxation changes dramatically when replacing the substitutional donor. Whereas central-cell effects induce marginal corrections for mobility [29–31], they are indispensable for spin relaxation in multivalley materials.

Figure 2(c) shows the temperature dependence of the spin lifetime in 10^{19} cm^{-3} Si:P (solid line). The calculation considers both electron-impurity and electron-phonon interactions. The latter has already been quantified [47,48], and was shown to be dominated by an intervalley f process due to scattering with shortwave Σ -axis phonons [49,50]. Also shown are contributions from electron-phonon intravalley and g -process scattering due to interactions with the crystal deformation potential and Δ -axis phonons, respectively [49]. We find that intravalley scattering off the potential tail of ionized impurities, which largely sets the mobility in doped semiconductors, is responsible for orders of magnitude weaker spin relaxation than all shown mechanisms in Fig. 2(c) [37]. The f processes do not vanish at the lowest order and dominate the spin relaxation which at low temperatures comes from scattering off the impurity central cell and at elevated temperatures from scattering with Σ -axis phonons. On the other hand, intravalley and g -process spin-flip scattering are weaker effects due to space inversion and time reversal symmetries [37,51]. Figure 2(d) shows the dependence of spin relaxation on doping concentration in Si:As. The spin relaxation enhancement is evident when entering the metallic regime ($N_d > 2 \times 10^{18} \text{ cm}^{-3}$) due to a change in the f -process dominant mechanism from electron scattering with Σ -axis phonons to electron scattering off the central-cell potential of impurities.

A novel aspect of the central-cell effect is the governing role of Yafet rather than Elliott spin-flip processes; that is, the spin relaxation is driven by the SOC of impurity scatterers rather than that of host-material atoms. The Elliott spin-flip process takes into account only the spin mixing of states from the host SOC, resulting in dopant-independent spin relaxation in contrast to the empirical trend shown in Fig. 2(a) [but similar to the mobility trend shown in Fig. 2(b)]. Similar to the study of momentum scattering [29,31], the Elliott process only probes the spinless part of the core potential [52], and therefore cannot account for the pronounced dopant-type dependence of the spin relaxation. We repeat our analysis for n -type germanium (Ge), which has four valleys centered at the L point of the Brillouin zone not connected by time reversal. The valley-orbit interaction in the central cell splits the $1s$ donor state in Ge to spin-independent singly and triply degenerate states [34]. The latter is further split by the SOC and this splitting is the only relevant energy scale for impurity-induced spin relaxation (detailed derivation is provided in the Supplemental Material [37]). A similar role of the

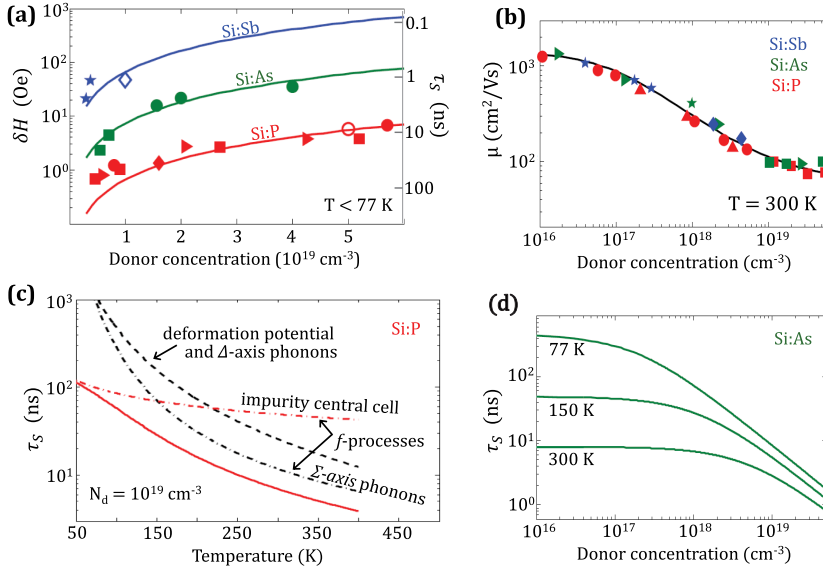


FIG. 2 (color online). (a) Spin relaxation in heavily doped n -type Si for three common donor types, {P, As, Sb}, at low temperature. Solid lines denote the theory results for the average spin lifetime (right axis), and symbols denote empirical values of the measured linewidth in EPR and spin injection experiments (left axis, open circle [15], open diamond [39], filled diamond [18], forward triangle [19], filled square [20], filled circle [21], star [22]). The scales on the left and right axes are related by $\delta H = 1/\gamma_e \tau_s$ where $\gamma_e = 1.7 \times 10^7 \text{ s}^{-1}$. Oe^{-1} is the electron gyromagnetic ratio. (b) Room-temperature mobility versus donor concentration showing a marginal dependence on donor identity (filled square [30], forward triangle [42], star [43], filled diamond [44], upward triangle [45], filled circle [46]). (c) Temperature dependence of τ_s in 10^{19} cm^{-3} Si:P (solid line). Intervalley f processes dominate the relaxation at all temperatures. (d) Doping concentration dependence of τ_s in Si:As at three temperatures. The decay of τ_s at the metallic regime ($N_d > 2 \times 10^{18} \text{ cm}^{-3}$) is due to transition from electron-phonon to electron-impurity dominated relaxation.

central-cell SOC is identified, in addition to a previously overlooked zeroth-order Elliott contribution to the intervalley spin flip in Ge.

Ramifications of the studied central-cell-driven spin relaxation extend beyond diamond structure crystals. The prerequisite condition for a strong signature of the effect is that conduction electrons populate k space regions that cannot be connected by time reversal ($\mathbf{k} \rightarrow -\mathbf{k}$). This condition is satisfied by most known metals where the Fermi surfaces extend across the Brillouin zone, by rock salt crystals such as lead telluride (PbTe) whose conduction-band edge is at the L points, or by complex oxides such as strontium titanate (SrTiO_3) whose conduction band along the Γ - X direction is flat due to its d -orbital nature. This condition does not apply in materials where thermal electrons populate a single zone-center valley such as in GaAs or if all the distinct valleys are related by time reversal such as in graphene. In these cases, spin flips due to short-range impurity scattering vanish in the lowest order (as we showed, for the vanishing “ g -process” spin flip due to time reversal symmetry, and for intravalley scattering due to the rotation operation around the high symmetry valley axis). Of all materials that obey the prerequisite condition, one should focus on crystals that respect space inversion symmetry. In multivalley materials that lack a space inversion center such as AIAs or GaP semiconductors with zinc-blende crystal structure, the Dyakonov-Perel spin relaxation mechanism can compete with the studied spin-flip effect at elevated temperatures [53]. In addition, one should also focus on materials whose Fermi surfaces exclude spin hot spots (regions in k space where band degeneracy is lifted by the SOC). Spin hot spots appear, for example, in small

regions of the Fermi surface of aluminum and can dominate its spin relaxation due to strong spin mixing of states in these regions [54]. These spin hot spots also explain the ultrashort spin lifetime of holes in unstrained bulk sp^3 semiconductors [55–57]. In these semiconductors, the SOC lifts the sixfold band degeneracy at the top of the valence band, rendering a strong spin mixing of hole states.

In conclusion, we have identified a general spin relaxation mechanism in multivalley materials whose understanding fills a long-standing gap in the theory of spin relaxation mechanisms in n -type semiconductors. The new formalism is valuable for characterization of silicon-based spintronic devices. For example, it can be used to optimize on-chip spin communications over millimeter length scales [2,58,59]. Knowing that the intervalley f process dominates the spin relaxation, one can prolong the spin lifetime by lifting the valley degeneracy [2,60,61]. Application of uniaxial compressive strain in Si along the [001] crystallographic direction raises the energies of perpendicular valleys relative to the two valleys whose axis is along the stress direction. As a result, the four valleys of higher energy are depopulated and, if the valley splitting is large compared with $k_B T$, electrons experience neither elastic nor inelastic f -process scattering via impurities or shortwave phonons, respectively. Finally, lifting the valley degeneracy in Ge by strain along the [111] crystallographic axis can lead to an exceptionally long spin lifetime given the absence of g processes and ultraweak intravalley spin flips in this material [10,62].

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