

Spin relaxation of electrons and holes in zinc-blende semiconductors

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We develop a procedure to calculate spin relaxation times of electrons and holes in semiconductors using full band structures. The spin-orbit (SO) interaction is included in the unperturbed Hamiltonian. With the use of spin projection operators, we calculate electron and hole spin relaxation from both Elliott-Yafet and D'yakonov-Perel' mechanisms, and quantitatively explain measurements of GaAs. The predicted relaxation times of GaN are longer for electrons, but shorter for holes. We find that the valence band SO splitting at the zone center is not a good indicator of SO coupling for electrons.

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A comprehensive understanding of spin relaxation of electrons and holes is of great scientific and technological importance, particularly in designing devices to exploit the spin degree of freedom of free carriers.^{1,2} Although considerable understanding of electron spin relaxation has been achieved,³⁻⁸ there has been no corresponding treatment of holes. Understanding spin relaxation of holes is particularly important as most magnetic semiconductors with high Curie temperatures to date are *p* type.⁹ Recent measurements indicate a very short (~ 0.1 ps) spin relaxation time for holes in GaAs.¹⁰

In this paper, we develop a physically intuitive approach to study spin relaxation limited by the Elliott-Yafet (EY)¹¹ and the D'yakonov-Perel' (DP)¹² mechanisms for *both* electrons and holes. Our approach incorporates these spin relaxation mechanisms with an accurate, nonperturbative treatment of the spin-orbit (SO) coupling. It has been shown that accurate energy bands are required for quantitative interpretation of the measured electron spin relaxation times.³⁻⁸ This is even more important for holes because simplified models usually do not adequately describe the heavily anisotropic valence bands. The calculated electron and hole spin relaxation times in GaAs are in good agreement with experiments.¹⁰⁻¹³ We also predict longer electron spin relaxation times in zinc-blende GaN,⁸ but shorter spin relaxation times for holes. We find, contrary to common interpretation, that the splitting between the heavy-hole and the split-off bands at the zone center (Δ_{SO}) is not always an accurate measure of the SO coupling strength for electrons.

The general Hamiltonian of a bulk semiconductor contains the orbital term (H_0), the SO interaction (H_{SO}), and spin-independent scattering (H_{sc}). Unlike many previous studies,^{5-8,11,12} where both H_{SO} and H_{sc} are treated as perturbations to H_0 , we include the SO interaction in H_0 and treat scattering by ionized impurities and phonons (H_{sc}) as a perturbation. Although the SO interaction was treated nonperturbatively before in spin lifetime calculations in zinc-blende semiconductors³ and in metals,¹⁴ only the DP³ or the EY mechanism¹⁴ was considered. The unperturbed energy bands are obtained by diagonalizing

$\tilde{H}_0 (=H_0+H_{SO})$, $\tilde{H}_0|n\mathbf{k}\sigma\rangle = E_{n\mathbf{k}\sigma}|n\mathbf{k}\sigma\rangle$. Here n is the band index, \mathbf{k} is the wave vector, and $\sigma(=1,2)$ is the (pseudo)spin label. Because of the SO interaction, the (pseudo)spin orientation depends on \mathbf{k} . The inclusion of H_{SO} in \tilde{H}_0 also leads to a \mathbf{k} -dependent energy splitting $\Delta E(n\mathbf{k}) \equiv |E_{n\mathbf{k}1} - E_{n\mathbf{k}2}|$.

The central issue to calculate spin relaxation using eigenstates $|n\mathbf{k}\sigma\rangle$ is to establish the relationship between the spin relaxation rate and the scattering matrix elements $\langle n'\mathbf{k}'\sigma_2|H_{sc}|n\mathbf{k}\sigma_1\rangle$. This can be achieved by using the density matrix and projection operators in spin space. Since the H_{SO} is included in the unperturbed Hamiltonian, spin is no longer a good quantum number and the spin polarization of a system should be described by an expectation value over the carrier distribution, $s = \text{Tr}(\hat{\rho}\hat{\sigma})/2$, where $\hat{\rho}$ and $\hat{\sigma}$ are the (electron or hole) spin density and Pauli matrices, respectively. Using the two eigenstates at band n with wave vector \mathbf{k} , we can construct the following 2×2 density matrix in spin space for electrons or holes:

$$\hat{\rho}^{\uparrow(\downarrow)}(n\mathbf{k}) = \sum_{\sigma\sigma'} \rho_{\sigma\sigma'}^{\uparrow(\downarrow)}(n\mathbf{k}) |n\mathbf{k}\sigma\rangle \langle n\mathbf{k}\sigma'|, \quad (1)$$

$$\hat{\rho}_{\sigma\sigma'}^{\uparrow(\downarrow)}(n\mathbf{k}) = \langle n\mathbf{k}\sigma | \hat{u}_{\uparrow(\downarrow)}^\dagger \hat{u}_{\uparrow(\downarrow)} | n\mathbf{k}\sigma' \rangle, \quad (2)$$

where $\hat{u}_{\uparrow(\downarrow)} \equiv [1 + (-)\hat{\sigma} \cdot \mathbf{m}]/2$ is the up-spin (down-spin) projection operator onto the quantization axis \mathbf{m} [chosen to be $(0, 0, 1)$]. These density matrices satisfy the normalization condition $[\text{Tr} \hat{\rho}^{\uparrow(\downarrow)}(n\mathbf{k}) = 1]$ and the completeness condition $[\hat{\rho}^\uparrow(n\mathbf{k}) + \hat{\rho}^\downarrow(n\mathbf{k}) = \hat{\mathbf{1}}]$ where $\hat{\mathbf{1}}$ is the unit matrix in spin space. For a state whose wave function cannot be factored into a spin part and an orbital part, the magnitude of spin expectation value for $\hat{\rho}^{\uparrow(\downarrow)}$ would be smaller than 1/2.

Since spins in different eigenstates $|n\mathbf{k}\sigma\rangle$ point along different directions, scattering between these eigenstates does not contribute to spin relaxation equally. The EY mechanism originates from spin-flip scatterings,¹¹ whereas the DP mechanism originates from spin precession accompanied by

spin-conserving scatterings.¹² These spin-flip and spin-conserving scatterings from $n\mathbf{k}$ to $n'\mathbf{k}'$ can be obtained by using the density matrices described above

$$w_{n\mathbf{k} \rightarrow n'\mathbf{k}'}^{\text{flip(cons)}} = \frac{2\pi}{\hbar} \sum_{\sigma_1 \sigma_2 \sigma_1' \sigma_2'} \rho_{\sigma_1' \sigma_2'}^{(\dagger)}(n'\mathbf{k}') \langle n'\mathbf{k}' \sigma_1' \sigma_2' | H_{sc} | n\mathbf{k} \sigma_1 \rangle \times \rho_{\sigma_1 \sigma_2}^\dagger(n\mathbf{k}) \langle n\mathbf{k} \sigma_2 | H_{sc}^\dagger | n'\mathbf{k}' \sigma_2' \rangle g(E_{n\mathbf{k}} - E_{n'\mathbf{k}'}), \quad (3)$$

where $g(E) = \delta(E)$ for an elastic scattering and $g(E) = \delta(E \pm \hbar\omega)$ for an inelastic scattering involving a phonon absorption/emission with ω being the optical phonon frequency. $E_{n\mathbf{k}} = \rho_{11}^\dagger E_{n\mathbf{k}1} + \rho_{22}^\dagger E_{n\mathbf{k}2}$.

It is straightforward to obtain the spin relaxation time of an electron or hole at band n with a given energy $E_{n\mathbf{k}}$ due to the EY mechanism from

$$\frac{1}{\tau_s^{EY}(E_{n\mathbf{k}})} = \sum_{n'\mathbf{k}'} w_{n\mathbf{k} \rightarrow n'\mathbf{k}'}^{\text{flip}}. \quad (4)$$

To calculate the spin relaxation time limited by the DP mechanism, we need to determine the spin precession vector $\Omega(n\mathbf{k})$ which is directly related to the spin splitting, $\Delta E(n\mathbf{k}) = \frac{1}{2}\hbar\Omega(n\mathbf{k}) \cdot \hat{\sigma}$.^{3,12} The amplitude $|\Omega(n\mathbf{k})|$ is $1/\hbar|E_{n\mathbf{k}1} - E_{n\mathbf{k}2}|$. Following steps similar to that of Ref. 12, we get

$$\frac{1}{\tau_s^{DP}(E_{n\mathbf{k}})} = \frac{2}{3} \bar{\Omega}^2 \left\{ \sum_{\mathbf{k}'} w_{n\mathbf{k} \rightarrow n\mathbf{k}'}^{\text{cons}} [1 - P_3(\mu)] \right\}^{-1}, \quad (5)$$

where P_3 is the third order Legendre polynomial, $\mu = \mathbf{k} \cdot \mathbf{k}' / |\mathbf{k}||\mathbf{k}'|$ and $\bar{\Omega}^2 = \int \Omega^2(n\mathbf{k}) d\omega_k / 4\pi$. We approximate $|\Omega|^2$ by the $l=3$ term of an expansion of Ω in spherical harmonics. Note from Eq. (5) that the spin-conserving scattering must be within the same band for the DP mechanism. This leads to an important observation that the DP mechanism does not contribute to spin relaxation of heavy holes, as that would require spin precession from $m_j = \pm 3/2$ to $m_j = \mp 3/2$, which is forbidden because $|\Delta m_j| > 1$. We emphasize that Eq. (5) is only valid when $|\Omega|\tau_p \ll 1$ (τ_p is the momentum relaxation time). In the opposite limit, the spin relaxation time is controlled simply by the precession operator, $1/\tau_s^{DP} \approx |\hat{\Omega}|$. In either case, the total spin relaxation time for a state $n\mathbf{k}$ is

$$1/\tau_s(E_{n\mathbf{k}}) = 1/\tau_s^{EY}(E_{n\mathbf{k}}) + 1/\tau_s^{DP}(E_{n\mathbf{k}}). \quad (6)$$

The above equations enable a direct computation of the carrier spin lifetimes from accurate electronic structures obtained from various approaches without relying on the formalism in Ref. 12 that was based on perturbation and a simplified band structure.

For H_0 we adopt an accurate tight-binding Hamiltonian that combines long-range tight-binding (sp^3 basis) and empirical pseudopotentials.¹⁵ Its parameters were fitted to a recently developed self-consistent GW theory, which as will be shown elsewhere,¹⁶ predicts quasiparticle levels (~ 0.2 eV) and effective masses to a high degree of accuracy for broad classes of materials, including III-V and II-VI compounds.

TABLE I. Spin-orbit couplings, LO phonon energies, and donor ionization energies in units of milli-electron volt and hole effective masses for GaAs and GaN. The masses are calculated from the first-principles band structures.

	λ_c	λ_a	Δ_{SO}	$\hbar\omega_{LO}$	E_D	m_{LH}^{100}	m_{LH}^{111}	m_{HH}^{100}	m_{HH}^{111}
GaAs	150	360	349	36.2	5.8	0.089	0.079	0.380	0.847
GaN	150	6	17.7	88.8	17.0	0.22	0.20	0.88	3.1

The spin-orbit interaction $\lambda\mathbf{L} \cdot \mathbf{S}$ is included in the on-site Hamiltonian of the sp^3 basis with two coupling parameters, one for cation (λ_c) and the other for anion (λ_a). The values of λ_c and λ_a are chosen to reproduce Δ_{SO} obtained from the first-principles GW theory (Table I).^{15,16} For H_{sc} we include scattering from ionized dopants¹⁷ and LO phonons. In our calculations we use the widely accepted donor ionization energies, E_D , and LO phonon energies, $\hbar\omega_{LO}$, for GaAs and GaN,¹⁸⁻²⁰ which are listed in Table I. We consider in this work only zinc-blende GaN, although wurtzite GaN is more commonly synthesized.

The calculated electron spin relaxation times due to the EY and DP mechanisms as a function of the electron energy at room temperature are shown in Fig. 1. The EY mechanism is important only at very low energy (≤ 30 meV), and DP dominates at higher energies. At the energy threshold for phonon emission, the momentum scattering dramatically increases, and consequently the spin relaxation time limited by the DP mechanism ($\tau_s^{-1} \sim \bar{\Omega}^2 \tau_p$) also increases abruptly. Similarly, scattering by impurities and phonons is weaker at low temperatures than that at high temperatures, leading to shorter spin relaxation times in the DP regime. After the phonon emission threshold, the momentum relaxation time is not very sensitive to the temperature and the kinetic energy.⁸ In this regime the spin relaxation limited by DP mechanism

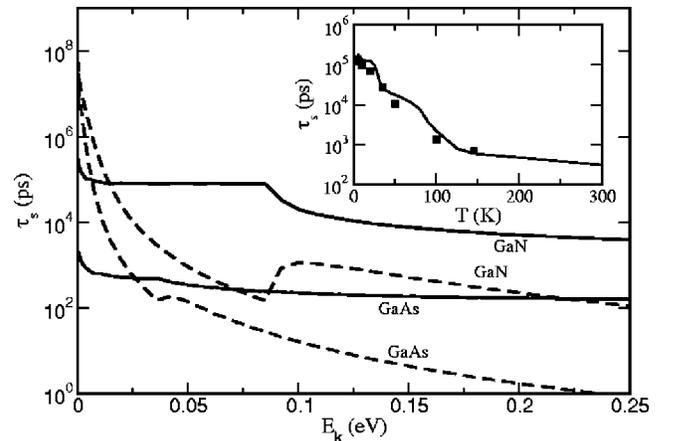


FIG. 1. Electron spin relaxation times due to the EY and DP mechanisms as a function of the electron energy in GaAs and GaN at $T=300$ K. Solid and dashed lines correspond to the contribution from the EY mechanism and the DP mechanism, respectively. The doping concentration is 10^{16} cm^{-3} and a photogenerated carrier density of $2 \times 10^{14} \text{ cm}^{-3}$ is added to the system. The inset plots the averaged spin relaxation time as a function of temperature in GaAs. Squares are experimental data reported in Ref. 13.

is determined by \mathbf{k} -dependent $\Omega(n\mathbf{k})$, which increases with energy, and therefore the spin relaxation time decreases with energy. Here the electron energy relaxation is neglected. In the inset of Fig. 1, we plot the temperature-dependent spin relaxation times in GaAs averaged over the distribution function at different temperatures. The calculated results quantitatively agree with experimental measurements.¹³ We note that our results for GaN are quite different from experiments.²¹ While it is possible that the sample in Ref. 21 is wurtzite GaN, which has a different band structure than that of zinc-blende GaN and may give rise to different spin relaxation times, the low carrier mobility and large threading dislocation density suggest that the GaN sample is in the metal-insulator-transition regime, where our theory is not applicable.²²

The numerical results obtained here for GaAs agree reasonably well with previous spin relaxation time calculations,⁸ where the spin-independent full-band structure is used to obtain the momentum relaxation rates but the spin scattering was included perturbatively using the simplified $k\cdot p$ formalism in Ref. 12. However, for GaN the electron spin relaxation times predicted in the present calculations are smaller by two orders of magnitude than those obtained in the perturbative approach. This difference arises from an underestimate of the SO interaction for the conduction electrons in GaN by using the Δ_{SO} in the commonly used simplified (eight-band) $k\cdot p$ formalism, although we emphasize that more elaborate (e.g., 14-band or 20-band) $k\cdot p$ schemes provide more accurate band structure description.³ In the simplified $k\cdot p$ formalism, the spin splitting in the conduction band of zinc-blende semiconductors due to the spin-orbit coupling is¹²

$$\Delta E(\mathbf{k}) = \alpha_c \hbar^3 (2m_e^3 E_g)^{-1/2} |\kappa|, \quad (7)$$

where $\kappa_z = k_z(k_x^2 - k_y^2)$, with κ_x and κ_y obtained by cyclic permutation, m_e is the electron effective mass, and E_g is the band gap. The coefficient $\alpha_c = 4m_e\eta/3m_{cv}(1-\eta/3)^{1/2}$ and $\eta = \Delta_{SO}/(E_g + \Delta_{SO})$. The value of m_{cv} is usually chosen to be $m_0/\sqrt{3}$, where m_0 is free electron mass.^{6,8,12} Figures 2(a) and 2(b) compare the k -dependent spin splitting for k_{\parallel} [110] obtained from our full-band structures with that predicted by Eq. (7). For GaAs we see that the two models agree well at small \mathbf{k} ($|\mathbf{k}a/2\pi| \leq 0.05$). The difference increases progressively with $|\mathbf{k}|$, resulting in about 100% increase near $|\mathbf{k}a/2\pi| = 0.1$. Notice that in GaN, the spin splitting predicted by Eq. (7) even near the Γ point is only about 15% of that obtained from the full-band structure. Consequently, spin scattering in GaN is dramatically underestimated when the simplified $k\cdot p$ formalism is used. According to our full-band structure, $\Delta_{SO} = f_c^p \lambda_c + f_a^p \lambda_a$, where f_c^p and $f_a^p (= 1 - f_c^p)$ are, respectively, cation (Ga) and anion (N or As) p composition of the valence wave function at the Γ point. The Δ_{SO} in GaN is small (17 meV) because the valence band is anion rich [see Fig. 2(c)] and λ_N is only about 6 meV. However, the conduction band wave functions are cation rich [see Fig. 2(c)] and λ_{Ga} is large (about 150 meV), resulting in a larger effective spin splitting in GaN. Thus in nitrides where Δ_{SO} is small, the simplified $k\cdot p$ formalism tends to underestimate

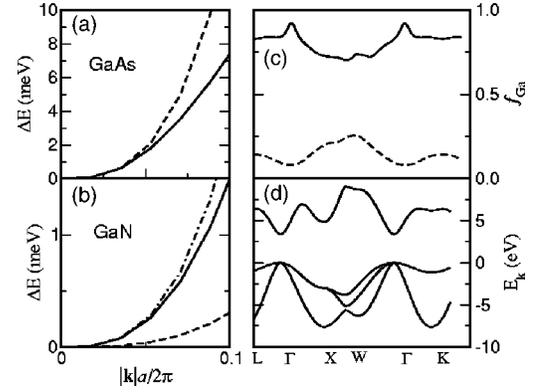


FIG. 2. The spin splitting of conduction band as a function of wave vector along [110] in (a) GaAs and (b) GaN. Solid and dashed lines represent the spin splitting obtained from the full-band structure and from Eq. (7), respectively. The dot-dashed line is a fit obtained by using $m_{cv} = 0.092m_0$ in Eq. (7) for GaN. (c) The cation (Ga) composition in the conduction band (solid line) and the heavy-hole band (dashed line) of GaN. (d) The full-band structure of GaN.

the SO interaction in the conduction band, and in systems where the anion SO coupling is very strong, such as GaSb, the simplified $k\cdot p$ formalism tends to overestimate the SO interaction in the conduction band. We see that Δ_{SO} alone is not a good measure of electron SO coupling. However, since the simplified $k\cdot p$ formalism regards m_{cv} as an adjustable parameter, $\Delta E(\mathbf{k})$ at small \mathbf{k} from Eq. (7) can be fit to its full-band structure value by an appropriate value of m_{cv} , as illustrated by the dot-dashed line in Fig. 2(b). We find such value for GaN to be $0.092 m_0$, considerably smaller than the most commonly used value, $m_0/\sqrt{3}$.

The approach developed here applies equally well in the studies of hole spin scattering. However, because of possible scattering between heavy-hole, light-hole, and spin-orbit bands, the study of hole scattering is much more complicated than that of electrons. The calculated spin relaxation times of heavy holes in GaAs and GaN as functions of the hole energy at two different temperatures are shown in Fig. 3. The EY mechanism is the sole origin for the heavy-hole spin relaxation and the dominant one for the light-hole spin relaxation. The much shorter spin relaxation times of holes than those of electrons can be understood by noticing that in any state of the light-hole and the split-off bands, up-spin and down-spin are strongly mixed, which is in sharp contrast to the situation of conduction-band states, where the mixture between up-spin and down-spin is usually very small. Thus when a heavy hole scatters into a light-hole state or vice versa, a significant amount of spin will flip, giving rise to very fast spin relaxation.

We see that the room temperature spin relaxation time of heavy holes in GaAs is about 0.1 ps, and it is not sensitive to the temperature or the kinetic energy. This value is in good agreement with the recent measurements of hole spin relaxation times in GaAs.¹⁰ The spin relaxation time at 5 K is only slightly longer than that at 300 K. However, we see that the spin relaxation time of heavy holes in GaN is an order of magnitude shorter than that in GaAs. The reason for the reduction is threefold. First, because of smaller Δ_{SO} in GaN,

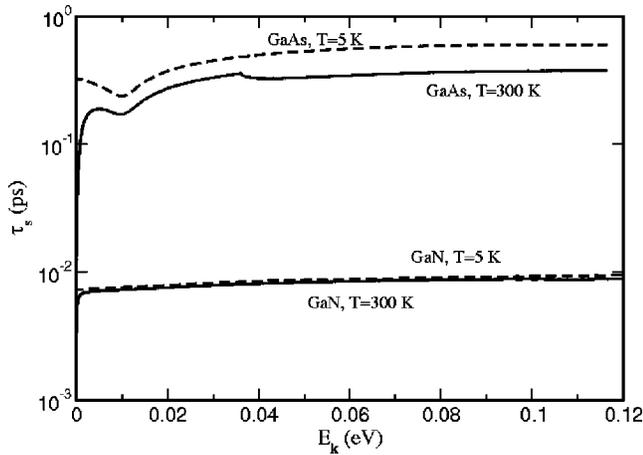


FIG. 3. Energy-dependent heavy hole spin relaxation time in 10^{16} cm^{-3} n -doped GaAs and GaN at 5 K (dashed lines) and 300 K (solid lines).

both the light-hole and the split-off bands are accessible for the heavy holes to scatter into. Second, the first-principles¹⁶ and other band-structure calculations²⁰ indicate that the effective mass of holes, and accordingly the density of states of the valence band in GaN, is larger than that in GaAs (Table I). Third, in spite of large difference in the effective mass of heavy holes and light holes, in GaN, the bands obtained in first principles have nearly the same dispersion everywhere in the Brillouin zone, except very close to Γ ^{16,20} due to the warping of the light-hole band in the presence of the SO interaction. Figure 2(d) shows the full-band structure of GaN. This degeneracy enables even the forward scatterings ($|\mathbf{k}' - \mathbf{k}| \sim 0$) between the two bands to flip spin. Therefore, spin relaxation due to the EY mechanism, the only channel for heavy holes, is proportionally larger in GaN. Since the LO phonon emission by a heavy hole requires large momentum change, which results in small scattering matrix elements, there is no apparent peak or valley in the spin relaxation time at the LO phonon energy. The enhanced forward scattering also leads to a very weak temperature dependence of spin relaxation, as shown in Fig. 3.

The calculated spin relaxation times of light holes are shown in Fig. 4. In spite of smaller effective mass, spin relaxation times of light holes in GaAs are about 0.1 ps, slightly shorter than those of heavy holes. The shorter spin relaxation times are due to an additional channel available for the light holes through the DP mechanism between

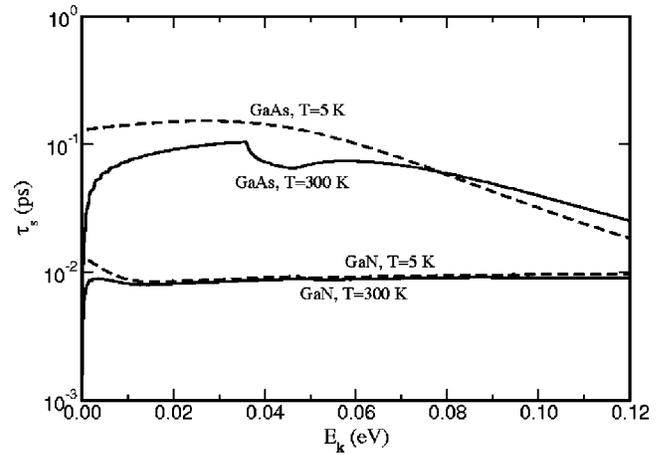


FIG. 4. Energy-dependent light hole spin relaxation time in 10^{16} cm^{-3} n -doped GaAs and GaN at 5 K (dashed lines) and 300 K (solid lines).

$J_m = \pm 1/2$ states. For GaN the light-hole spin relaxation times are essentially identical to the heavy-hole spin relaxation times, resulting from the nearly degenerate heavy-hole and light-hole bands.

In summary, we have developed a systematic approach to study spin relaxation limited by the EY and the DP mechanisms for both electrons and holes. Salient features of this approach are that (a) full-band structures are used, (b) the SO interaction is included nonperturbatively, and (c) electron and hole spin relaxation due to both the EY and the DP mechanisms is taken into account. We applied this approach to study spin relaxation in zinc-blende GaAs and GaN. Our numerical calculation of spin relaxation times in GaAs explains the measured values. We predict an order-of-magnitude shorter hole spin relaxation times and two orders-of-magnitude longer electron spin relaxation times in high-quality samples of GaN than those in GaAs. We also showed that Δ_{SO} alone is not a reliable estimate of the SO interaction for conduction electrons. A more accurate description requires two SO interaction parameters, one each for anion and cation. We conclude that electron spin relaxation times will be longer in materials with smaller cation SO parameters. Furthermore, longer hole spin relaxation times are possible only when the SO splitting at Γ is large.

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