Electron spin dynamics in GaN

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Gallium nitride is a promising material system for spintronics, offering long spin relaxation times and prospects for room-temperature ferromagnetism. We review the electron spin dynamics in bulk GaN. Time-resolved magneto-optical studies of both the wurtzite and the cubic phase of GaN show the dominance of Dyakonov–Perel (DP) relaxation for free conduction band electrons. Spin relaxation in the wurtzite phase is characterized by an intrinsic spin relaxation anisotropy and the limitation of spin lifetimes by a strong Rashba term. Spin lifetimes are strongly enhanced in cubic GaN, where only a weak Dresselhaus term contributes to DP relaxation. Ion-implanted wurtzite GaN shows a strong increase of electron spin lifetimes for increasing implantation dose, caused by increasing localization of carriers. The spin dynamics of conduction band electrons in Gd-implanted GaN as a candidate for a room-temperature ferromagnetic semiconductor is also only governed by localization effects and does not show signs of an efficient exchange coupling between the electrons and the magnetic Gd ions.

1 Introduction Spintronics as the novel spin-based semiconductor electronics has been an intense and fruitful field of research in the last years, leading to the discovery and detailed understanding of numerous spin effects [1–5]. In addition, a plethora of possible applications including, e.g., the famous spin transistor [6] or spin optoelectronic devices [7–9], has been proposed. However, for the realization of the proposed concepts challenging requirements are imposed on the underlying semiconductor materials, like, e.g., very long spin lifetimes or ferromagnetic behavior with Curie temperatures above room temperature. These challenges have triggered intense research on optimized structures and new materials. In particular, GaN has attracted strong interest as it promises to combine fascinating properties, ranging from weak spin–orbit coupling due to its light constituting elements over high-temperature ferromagnetism upon doping with rare earth ions [10] up to its good optoelectronic properties [11] and its well established use in the semiconductor industry [12]. In addition, GaN is a highly instructive model system for the influence of crystal symmetry on the electron spin dynamics as besides the thermodynamically stable wurtzite phase also the metastable cubic phase can be prepared [13]. While the spin dynamics in the dilute nitrides GaNAs and GaInNAs [14–18], and of holes [19] and especially excitons in GaN [20–23] has been intensively studied, the electron spin dynamics in GaN had been only rarely studied after the early work of Beschoten [24]. Here, we will give a review of our experimental investigations of the electron spin dynamics in bulk GaN by time-resolved magneto-optical Kerr-rotation (TRKR) spectroscopy [25–32]. After a short overview of the theory of electron spin relaxation in GaN, we will first discuss the spin dynamics in bulk wurtzite GaN in dependence on external magnetic field, temperature, and doping density. These results will afterwards be contrasted with the spin dynamics in bulk cubic GaN, before we conclude with a survey of the electron spin dynamics in ion-implanted GaN with special emphasis on Gd-implanted GaN as a candidate for a ferromagnetic semiconductor.

2 Theory of electron spin relaxation in GaN The theoretical description of electron spin relaxation in GaN can be approximately divided into two regimes for the purpose of this review, with the two limiting cases of spin relaxation of either free, delocalized electrons, or completely localized electrons.

2.1 Dyakonov–Perel spin relaxation of free electrons Spin relaxation of free, delocalized conduction band electrons is governed by the Dyakonov–Perel (DP)
mechanism [33] in most III–V semiconductors [5]. DP relaxation is based on an intrinsic conduction band spin splitting due to spin–orbit coupling (SOC). This spin splitting can be formally described by the Hamiltonian

$$\hat{H}_{soc} = \frac{\hbar}{2} \mathbf{\Omega}(\mathbf{k}) \cdot \mathbf{\sigma}$$  \hspace{1cm} (1)

with $\mathbf{\sigma}$ as the vector of Pauli spin matrices. In analogy to the Zeeman Hamiltonian for electrons in an external magnetic field, $\mathbf{\Omega}(\mathbf{k})$ is readily interpreted as an effective, $\mathbf{k}$-dependent magnetic field, which acts on the electron spins and causes them to precess. Random momentum scattering leads to a fluctuating effective magnetic field $\mathbf{\Omega}(\mathbf{k})$ and thus to spin dephasing of an electron ensemble. The underlying SOC is strongly dependent on the symmetry of the crystal lattice, hence distinctly different effective magnetic fields $\mathbf{\Omega}(\mathbf{k})$ exist in the wurtzite and cubic phase of GaN. The conduction band spin splitting in the wurtzite phase arises from two contributions, the $\mathbf{k}$-linear Rashba term [34–37] and a cubic $k^3$-term [38], with the corresponding effective magnetic field [39, 26]

$$\mathbf{\Omega}_{wz}(\mathbf{k}) = \frac{2}{\hbar} \left( \begin{array}{ccc}
\gamma_c (bk_x^2 - k_z^2) k_y + \alpha_c k_y \\
-\gamma_c (bk_x^2 - k_z^2) k_y + \alpha_c k_y \\
0
\end{array} \right),$$  \hspace{1cm} (2)

where $\mathbf{e}_z \parallel [0001]$ (c-axis), $\mathbf{e}_x \parallel [1\bar{1}20]$, $\mathbf{e}_y \parallel [1\bar{1}00]$, and $k_x^2 = k_y^2 + k_z^2$. The parameters $\gamma_c$ and $b$ determine the strength of the cubic contribution, while $\alpha_c$ gives the size of the Rashba contribution. The corresponding values of the SOC parameters from tight-binding, first-principle, and $k \cdot p$ calculations, respectively, are listed in Table 1.

The Rashba contribution vanishes in the cubic phase of GaN due to its higher symmetry, and the effective magnetic field is only due to the cubic $k^3$-Dresselhaus term [40]

$$\mathbf{\Omega}_{zb}(\mathbf{k}) = \frac{2\gamma_{zb}}{\hbar} \left( \begin{array}{ccc}
k_y (k_x^2 - k_z^2) \\
k_x (k_x^2 - k_y^2) \\
k_z (k_x^2 - k_y^2)
\end{array} \right)$$  \hspace{1cm} (3)

with $\gamma_{zb}$ as the spin splitting constant for the cubic phase (see Table 1). Both effective magnetic fields $\mathbf{\Omega}_{wz}(\mathbf{k})$ for the wurtzite and $\mathbf{\Omega}_{zb}(\mathbf{k})$ for the cubic phase, respectively, are schematically visualized in Figure 1.

The tensor of spin relaxation rates $\gamma_{ij}$ is obtained from the effective magnetic field in the most simplistic form of DP relaxation theory [42] via

$$\gamma_{ij} = \frac{1}{2} \left( \delta_{ij} (\mathbf{\Omega}^2) - \langle \mathbf{\Omega}, \mathbf{\Omega} \rangle \right) \tau_p$$  \hspace{1cm} (4)

where $i, j = x, y, z$, and $\tau_p$ is the momentum scattering time and $\langle \ldots \rangle$ denotes averaging over the momentum distribution of electrons. Averaging over an isotropic angular distribution of $\mathbf{k}$ already gives the symmetry of the spin relaxation tensor

$$\gamma_{wz} = \begin{pmatrix}
y_{xx,wz} & 0 & 0 \\
0 & y_{xx,wz} & 0 \\
0 & 0 & 2y_{xx,wz}
\end{pmatrix}$$  \hspace{1cm} (5)

for the wurtzite phase and

$$\gamma_{zb} = \begin{pmatrix}
y_{xx,zb} & 0 & 0 \\
0 & y_{xx,zb} & 0 \\
0 & 0 & y_{xx,zb}
\end{pmatrix}$$  \hspace{1cm} (6)

for the cubic phase, respectively.

### 2.2 Spin relaxation of localized electrons

The localization of electrons leads to an effective suppression of DP relaxation as the bound states have zero average wave vectors. Instead, spin dephasing is usually governed by hyperfine interaction with nuclear spins of the lattice atoms [43, 44]. The corresponding spin dephasing time can be approximated by [45, 46]

$$\tau_{s,nuc} = \hbar \frac{3N}{2 \sum I_i (I_i + 1) A_i^2 y_i}$$  \hspace{1cm} (7)

where $N$ is the number of nuclei overlapping with the electron wavefunction, $A_i$ is the hyperfine constant, $I_i$ the nuclear spin and $y_i$ the abundance of isotope $i$. In external magnetic fields, additional spin dephasing arises from a distribution $\Delta g$ of

| GaN | $\gamma_c$ (in eV Å$^3$) | $b$ | $\alpha_c$ |
|-----|----------------|-----|--------|
| wurtzite | 0.32$^a$ | 3.959$^a$ | 9.0$^b$ |
| cubic | 0.51$^a$ | – | – |
|    | 0.84$^c$ | – | – |

$^a$ From Ref. [39];
$^b$ From Ref. [41];
$^c$ From Ref. [28].

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the electron Landé $g$ factor, which results in a magnetic field dependence [44]

$$
\tau_{s,\text{loc}}(B_{\text{ext}}) = \frac{1}{\sqrt{\left(1/\tau_{s,\text{loc}}(0)\right)^2 + \left[\Delta g\mu_B B_{\text{ext}}/\hbar\right]^2}}
$$

with a characteristic $\tau_{s,\text{loc}}(B_{\text{ext}}) \propto 1/B_{\text{ext}}$ dependence for large magnetic fields $B_{\text{ext}}$.

### 3 Spin dynamics in bulk wurtzite GaN

We will start the review of our experimental results with the electron spin dynamics in bulk wurtzite GaN, discussing first the influence of external magnetic fields before turning to the temperature and doping density dependence of spin relaxation. The bulk wurtzite GaN samples used for these studies were grown by molecular beam epitaxy (MBE) on Si(111) substrates, with one additional sample grown on a GaN template. The top GaN layers were n-doped with Si, with doping densities from $n_d = 5 \times 10^{15} \text{ cm}^{-3}$ up to $1.5 \times 10^{19} \text{ cm}^{-3}$. Detailed sample information and the exact layer structures can be found in Ref. [27].

#### 3.1 Anisotropy of spin relaxation

First, we discuss the magnetic field dependence of the electron spin dynamics. Such measurements are extremely useful as they allow, e.g., to map the symmetry of the spin relaxation tensor as was demonstrated, for example, in GaAs (110) quantum wells [47, 48]. In bulk wurtzite GaN, a slowdown of spin relaxation is found if an external magnetic field is applied, as can already be seen from the raw TRKR transients on a semilog scale (see Fig. 2), where the transients decay significantly slower if an external magnetic field is applied. The corresponding spin relaxation time shows a sudden increase from its zero-field value $\tau_{s,zw,0}$ to a substantially longer value $\tau_{s,zw}^B$, for $B_{\text{ext}} > 0$, which is then approximately constant up to $B_{\text{ext}} = 1\text{ T}$ (see Figs. 3 and 7). This peculiar magnetic field dependence directly reflects an intrinsic spin relaxation anisotropy of bulk wurtzite GaN [25]. As can already be seen from the general form of the spin relaxation tensor $\gamma_{zw}$ with $\gamma_{zz,zw} = 2\gamma_{xx,zw}$ (cf. Eq. (5)), spins pointing along the $z$-direction (along the c-axis) relax twice as fast as spins in the $x$-$y$-plane. Figure 4 gives an intuitive explanation of the observed anisotropy: spins pointing in $z$-direction (cf. Fig. 4a) are susceptible to both the $x$- and $y$-component of the fluctuating effective magnetic field $\Omega_{zw}(k)$, while spins perpendicular to the $z$-direction are only subject to either the $x$- or the $y$-component of $\Omega_{zw}(k)$ and thus relax slower (see Fig. 4b). In the experiment, the zero-field measurements yield the relaxation rate $\gamma_{zz,zw} = 1/\tau_{s,zw}^0$. If an external magnetic field $B_{\text{ext}}$ is applied in the $x$-$y$-plane, the spins precess around $B_{\text{ext}}$, and an averaged relaxation rate $\gamma_{zz,zw} = (\gamma_{zz,zw} + \gamma_{yy,zw})/2 = 1/\tau_{s,zw}^B$ is observed (cf. Fig. 4c). Since $\gamma_{zz,zw} = 2\gamma_{xx,zw}$ (cf. Eq. (5)), an
increase from $\tau^{0}_{zz,\text{wz}}$ to $\tau^{R}_{zz,\text{wz}} = 4/3 \tau^{0}_{zz,\text{wz}}$ for $B_{\text{ext}} > 0$ is expected, in very good agreement with the experiment (see Fig. 3). This anisotropy reflects the symmetry of SOC in wurtzite semiconductors and is a clear fingerprint of DP relaxation of electrons in bulk wurtzite GaN.

We note that the anisotropic spin relaxation leads for very small external magnetic fields $B_{\text{ext}}$ to a renormalization of the Larmor precession frequency $\omega_L$ as discussed in Refs. [25, 47].

### 3.2 Temperature and electron density dependence

While the magnetic field-dependent measurements presented in the previous section allowed to map the symmetry of the spin relaxation tensor, experiments on the temperature, and doping density dependence of spin relaxation yield information on the $k$-dependence of the spin relaxation tensor [26, 27, 32].

The electron spin relaxation time of moderately n-doped samples shows a monotonic decrease for increasing temperature (see Fig. 5). This temperature dependence can be quantitatively explained by DP relaxation [26]. To calculate the temperature dependence within DP theory, we evaluate Eq. (4) for the effective magnetic field $\Omega_{\alpha\beta}(k)$ according to Eq. (2), assuming a Boltzmann distribution for the electron momentum, an isotropic effective mass $m^*$ and neglecting a $k$-dependence of the SOC parameters. A spin relaxation rate

$$\gamma^{R}_{zz,\text{wz}} = \frac{4 \alpha e^2 m^* k_B T}{\hbar^2} \tau_p$$

follows taking only the Rashba contribution into account. For only the cubic contribution, the spin relaxation rate

$$\gamma^{D}_{zz,\text{wz}} = \frac{4(24 - 8b + 3b^2)\gamma^2_e (k_B T)^3 m^*}{\hbar^8} \tau_p$$

is obtained. The coexistence of both the Rashba and cubic contribution leads to an additional interference term

$$\gamma^\text{int}_{zz,\text{wz}} = 8\alpha e^2 \gamma_e (b - 4) m^* (k_B T)^2 \frac{\tau_p}{\hbar^6}$$

that had been neglected in wurtzite semiconductors [49, 50] though being well-known from asymmetric 2D zincblende structures [51]. The spin relaxation time $\tau^{0}_{zz,\text{wz}}$ is in very good agreement with the experimental data as shown in Fig. 5, assuming a constant momentum scattering time $\tau_p = 40 \text{fs}$ corresponding to a mobility of $\mu = \tau_p e/m^* \approx 350 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$, which is in the typical range of comparable structures [52]. These results clearly demonstrate that spin relaxation in n-type GaN is governed by DP relaxation. Comparing the spin relaxation times computed for only the Rashba and only the cubic contribution (see Fig. 6a), one clearly finds that DP relaxation in GaN is completely dominated by the Rashba contribution up to room temperature. The interference term also contributes only very weakly (cf. Fig. 6b). We note that the intrinsic spin relaxation anisotropy persists up to room temperature (see the magnetic field-dependent measurements in Fig. 7). The investigation of the $k$-dependence of the spin relaxation ten-

![Figure 5](https://example.com/figure5.png)

**Figure 5** Temperature dependence of the spin relaxation time $\tau_s$ in two moderately n-doped bulk wurtzite GaN samples. The triangles show the zero field spin relaxation times, while the circles and diamonds refer to $3/4$ times the averaged spin relaxation time in magnetic fields to account for the spin relaxation anisotropy of bulk wurtzite GaN. The solid lines show the spin relaxation times calculated by Dyakonov–Perel theory. (Reprinted from Ref. [26]. Copyright (2010) by the American Physical Society.)

![Figure 6](https://example.com/figure6.png)

**Figure 6** (a) Temperature dependence of the spin relaxation time $1/\gamma^{R}_{zz,\text{wz}}$ in bulk wurtzite GaN calculated by Dyakonov–Perel theory. Shown are also the spin relaxation time $1/\gamma^{0}_{zz,\text{wz}}$ for only the Rashba contribution and $1/\gamma^{D}_{zz,\text{wz}}$ for only the cubic contribution. Spin relaxation in bulk wurtzite GaN is clearly dominated by the Rashba contribution. (b) Calculated temperature dependence of the spin relaxation time with and without the interference term, showing that it adds only a minute correction.
Figure 7 Magnetic field dependence of the spin relaxation time $\tau_s$ of the two moderately n-doped bulk wurtzite GaN samples of Fig. 5 for different temperatures. The spin relaxation anisotropy with its characteristic increase of $\tau_s$ in an external magnetic field persists at all temperatures. (Reprinted from Ref. [26]. Copyright (2010) by the American Physical Society.)

Figure 8 Electron density dependence of the spin relaxation time $\tau_s$ at (a) $T = 80$ K and (b) $T = 293$ K in eleven bulk wurtzite GaN samples. The critical density $n_{D,c}$ for the maximum spin lifetime as predicted by Dyakonov–Perel theory is marked by the dashed lines. (Reprinted from Ref. [27]. Copyright (2011) by the American Physical Society.)

The spin relaxation time $\tau_s$ can be extended much deeper into $k$-space than accessible by temperature-dependent measurements by studying the density dependence of spin relaxation [27]. Figure 8 shows the spin relaxation time $\tau_{s,B}$ for eleven GaN samples covering a broad doping range from $n_D = 5 \times 10^{15}$ cm$^{-3}$ up to $1.5 \times 10^{19}$ cm$^{-3}$ with the highest doping level corresponding to an approximate Fermi temperature $T_F = E_F/k_B = 1200$ K. The spin relaxation time shows a non-monotonic density dependence with only a moderate decrease for the highest densities. This non-monotonic density dependence results from the interplay between the density dependence of the effective magnetic field average

$$\langle \Omega_{\text{eff},wz}^2 \rangle \equiv \frac{1}{2} \left( \delta_{ij} \langle \Omega_i^2 \rangle \right) = i, j = z$$

and of the momentum scattering time $\tau_p$, and can be understood in analogy to theoretical predictions for zincblende semiconductors by Jiang and Wu [53]. The ratio $T_F/T_c$ of lattice temperature $T$ and Fermi temperature $T_F$ divides two regimes: in the non-degenerate regime, where $T_F \ll T$, the effective magnetic field average $\langle \Omega_{\text{eff},wz}^2 \rangle$ is approximately independent of the doping density, while the momentum scattering time $\tau_p \propto n_D^{-1}$ is decreasing with increasing density $n_D$ due to the dominance of electron–impurity and electron–electron scattering [5, 53]. The spin relaxation time $\tau_s$ increases therefore with increasing $n_D$ in the non-degenerate regime. On the contrary, the effective magnetic field average $\langle \Omega_{\text{eff},wz}^2 \rangle$ increases with density $n_D$ in the degenerate regime ($T_F \gg T$), while the momentum scattering time $\tau_p \propto \text{const.}$ is approximately constant [4, 5], leading to a decrease of $\tau_s$ with $n_D$. Overall, the observed non-monotonic density dependence results, with the maximum spin relaxation time at the cross-over of the two regimes at the critical density $n_{D,c}$, where $T_F(n_{D,c}) = T$. The experimental densities for the maximum spin relaxation time show the expected shift toward higher densities for increasing lattice temperature $T$ and agree also very well with the calculated values of $n_{D,c} = 2.3 \times 10^{18}$ cm$^{-3}$ for $T_F = 80$ K and $n_{D,c} = 1.6 \times 10^{19}$ cm$^{-3}$ for $T_F = 293$ K.

$$y_{\text{eff},wz}^R = \frac{4\pi^3/\hbar^2}{3\gamma^2/\hbar^2} \rho_{p,\text{eff},wz} n_D^{3/4} \tau_p$$

(14)

with a $y_{\text{eff},wz}^R \propto n_D^2$ dependence follows for only the Rashba contribution. The spin relaxation rate

$$y_{\text{eff},wz}^D = \frac{12(3\hbar^2 - 8b + 24)\pi^4/\hbar^2}{35\hbar^2} n_D^{5/3} \tau_p$$

(15)
for only the cubic contribution shows the characteristic $\gamma_{\text{eff}, \text{w}}^2\propto n^2_D$ dependence well known from semiconductors with zincblende structure [54]. The additional determination of the momentum scattering time $\tau_p$ by van-der-Pauw transport experiments allows to compare the purely experimentally determined product $\tau_s \cdot \tau_p$ to the theoretically predicted effective magnetic field averages $1/\langle \Omega^2_{\text{eff}, \text{w}} \rangle$ based on the relation $\tau_s \cdot \tau_p = 1/\langle \Omega^2_{\text{eff}, \text{w}} \rangle$ following from Eq. (4). The excellent agreement of the experimental data for $\tau_s \cdot \tau_p$ to $1/\langle \Omega^2_{\text{eff}, \text{w}} \rangle$ predicted for a pure Rashba contribution clearly demonstrates that the Rashba term governs spin relaxation in bulk wurtzite GaN up to the highest investigated density of $1.5 \times 10^{20} \text{cm}^{-3}$ (see Fig. 9a and b). We note that the spin relaxation anisotropy persists up to the highest densities. A dominance of the cubic contribution can be expected only for even higher densities $n_D > 10^{20} \text{cm}^{-3}$ (see Fig. 9c).

In conclusion, the electron spin dynamics in bulk wurtzite GaN is governed by DP relaxation with a characteristic intrinsic spin relaxation anisotropy, where spins pointing along the $c$-axis relax twice as fast as spins perpendicular to this axis. The $k$-linear Rashba term completely dominates spin relaxation up to very high densities exceeding electron densities of $10^{20} \text{cm}^{-3}$.

4 Spin dynamics in bulk cubic GaN

The Rashba contribution that dominates spin relaxation in bulk wurtzite GaN vanishes in the cubic phase of GaN due to its higher symmetry. One might therefore wonder whether spin relaxation times are much longer in cubic GaN than in wurtzite GaN, as was predicted theoretically [55, 56] and indicated by first experimental results [57]. We studied this question in two cubic GaN samples grown by MBE on 3C-SiC (001) substrates. Sample details are given in Ref. [28]. The cubic GaN is highly n-doped with Si to a carrier concentration $n_D = 1 \times 10^{19} \text{cm}^{-3}$.

4.1 Spin relaxation in highly $n$-doped cubic GaN

Already the raw TRKR data shown in Fig. 10a with signals persisting longer than 2 ns indicate very long spin relaxation times despite the high doping level. In addition, the very good match between the slopes of the TRKR transients for zero magnetic field and $B_{\text{ext}} = 0.27 \text{T}$ indicates isotropic relaxation, as is expected for the higher symmetry of the cubic phase as compared to wurtzite GaN (cf. Eq. (6)). Correspondingly, the spin relaxation time shows no dependence on the external magnetic field (see Fig. 10b).

The temperature dependence of the spin relaxation time shown in Fig. 11 is characterized by two remarkable features: first, the overall spin relaxation times are very long, even exceeding 0.5 ns at room temperature despite the high

![Figure 9](https://example.com/f9.png)

**Figure 9** Comparison of the purely experimentally determined product $\tau_s \cdot \tau_p$ of the spin relaxation time $\tau_s$ and the momentum scattering time $\tau_p$ from van-der-Pauw transport measurements with the theoretically predicted effective magnetic field average $1/\langle \Omega^2_{\text{eff}, \text{w}} \rangle$ for the degenerate samples of Fig. 8 at (a) $T = 80 \text{K}$ and (b) $T = 293 \text{K}$. (c) Calculated electron density dependence of the total effective magnetic field average $1/\langle \Omega^2_{\text{eff}, \text{w}} \rangle$ and of the effective magnetic field averages $1/\langle \Omega^2_{\text{eff}, \text{w}} \rangle$ for only the Rashba contribution and $1/\langle \Omega^2_{\text{eff}, \text{w}} \rangle$ for only the cubic contribution, respectively. The Rashba contribution dominates spin relaxation in bulk wurtzite GaN up to densities of $10^{20} \text{cm}^{-3}$. (Reprinted from Ref. [27]. Copyright (2011) by the American Physical Society.)
doping density of $1 \times 10^{19}$ cm$^{-3}$. These times are 2 orders of magnitude longer than in GaAs at comparably high doping levels [54]. Second, the spin relaxation time shows only a very weak temperature dependence and is virtually constant for $T > 75$ K. The observed features are well explained by DP relaxation in the highly degenerate regime, and confirm an early prediction of Dyakonov and Perel [33].

The spin relaxation rate in the degenerate regime is calculated from Eq. (4) and the effective magnetic field $\mathbf{b}(\mathbf{k})$ according to Eq. (3) for cubic GaN by averaging over an isotropic angular distribution of $\mathbf{k}$ and replacing the absolute value $k$ by the Fermi wavevector $k_F = (3\pi^2 n_D)^{2/3}$. A spin relaxation rate

$$\gamma_{cz, zb}^D = \frac{96\pi^4 \gamma^D_{cz, zb} n_D^2 \tau_p}{35\hbar^2}$$ (16)

follows [28, 31]. This spin relaxation rate shows only a very weak temperature dependence due to the weak temperature dependence of both $n_D$ and $\tau_p$ [58]. Using the value $\gamma_{cz, zb} = 0.84$ eV Å$^{-1}$ of Table 1, a constant spin relaxation time $\tau_{zh}^D \approx 570$ ps follows for a momentum scattering time of 40 fs, in good agreement with the experimental data (see Fig. 11). We note that a second sample with a higher wurtzite content due to non-optimal growth conditions shows comparably long spin relaxation times [31], demonstrating the robustness of spin coherence against phase impurity at least in the degenerate regime.

In conclusion, cubic GaN shows very long spin coherence due to its very small spin splitting constant, making cubic GaN a very promising material for future spintronics applications.

5 Spin dynamics in ion-implanted GaN This review of the electron spin dynamics in GaN has so far been focused on DP relaxation of free, delocalized electrons under the influence of intrinsic material properties, like symmetry or strength of SOC, as well as of external parameters, like temperature or external magnetic field. In praxi, however, a disorder potential exists in a semiconductor crystal that also has an impact on the spin dynamics via, e.g., modification of the momentum scattering or localization of carriers. The influence of the disorder potential on the spin dynamics can be investigated via implantation of ions, where the implantation damage strongly alters the disorder potential. In addition, the spin dynamics in ion-implanted GaN is also of high relevance for spintronics after various experimental reports of ferromagnetism in ion-implanted GaN [59–61]. Especially, Gd-implanted GaN has attracted strong interest as a possible ferromagnetic semiconductor for, e.g., spin injectors [62, 63]. We will first discuss the impact of implantation of nonmagnetic ions on the spin relaxation in GaN, before we finally review our results on the electron spin dynamics in Gd-implanted GaN.

5.1 Ga and Au ion implanted wurtzite GaN The implantation of various doses of Ga ions into GaN allows to tune the disorder potential and to study its influence on the spin dynamics. For this purpose, we implanted different Ga$^+$ doses into a 1.8 μm thick GaN layer grown by metal-organic chemical vapor deposition (MOCVD) on a sapphire substrate. For comparison, also different doses of Au$^{2+}$ ions were implanted. The sample and the implantation procedure are in detail described in Ref. [29]. Figure 12a shows the

Figure 11 Temperature dependence of the spin relaxation time $\tau_s$ in the highly n-doped cubic GaN sample of Fig. 10. The weak temperature dependence of $\tau_s$ is in very good agreement with predictions of Dyakonov–Perel theory for the highly degenerate regime (see solid line). (Reprinted with permission from Ref. [28]. Copyright 2010, AIP Publishing LLC.)

Figure 12 (a) Spin relaxation times $\tau_{s,0}$ for zero magnetic field and $\tau_{s,B}$ for a moderate external magnetic field $B_{ext} = 0.2$ T at $T = 80$ K in bulk wurtzite GaN implanted with different Ga densities. (b) Ratio $\beta$ of the spin relaxation times $\tau_{s,0}$ and $\tau_{s,B}$ for the different implanted densities. The increase of both the spin relaxation times and of $\beta$ is caused by increasing electron localization at implantation defects. (Reprinted with permission from Ref. [29]. Copyright 2013, AIP Publishing LLC.)
spin relaxation time $\tau_{s,0}$ for zero magnetic field and $\tau_{s,B}$ for a moderate external magnetic field ($B_{ext} = 0.2$ T) for various implanted Ga doses. The observed strong increase of the spin relaxation time by more than an order of magnitude for increasing Ga dose is accompanied by a change of the magnetic field dependence (see Fig. 13), which can be quantified by the ratio $\beta = \tau_{s,0}/\tau_{s,B}$. The strong increase of the spin relaxation time for increasing implantation dose is caused by localization at implantation defects, which is sensitively probed by the ratio $\beta$. Free, delocalized electrons are subject to DP relaxation with the intrinsic spin relaxation anisotropy of wurtzite GaN as discussed in Section 3.1. The characteristic increase of the spin relaxation time from its zero-field value $\tau_{s,0}$ to $\tau_{s,B} = 4/3\tau_{s,0}$ in external magnetic fields leads to a value $\beta_{DP} = 0.75$ for DP relaxation of free electrons, which is observed for the lowest implantation densities (see Fig. 12b). For localized electrons, in contrast, a ratio $\beta_{loc} > 1$ follows due to the $\tau_{s,loc} \propto 1/B_{ext}$ magnetic field dependence as discussed in Section 2.2. The change of the magnetic field dependence from anisotropic DP relaxation for the lowest implantation dose (see Fig. 13b) to the $\tau_{s,B} \propto 1/B_{ext}$ dependence for the highest implanted dose (cf. Fig. 13c) and the corresponding increase of $\beta$ from $\beta = 0.75$ up to $\beta \approx 1.2$ (see Fig. 12b) directly witnesses the transition from delocalized electrons to electrons strongly localized at the defects created during implantation. The increasing localization also explains the observed slowdown of spin relaxation as the spin relaxation of localized electrons due to hyperfine interaction is less effective than the DP relaxation of delocalized electrons. The spin relaxation time $\tau_{s,loc} \approx 1.8$ ns estimated from Eq. (7) for completely localized electrons [29] is in very good agreement with the saturation value of the spin relaxation time for the highest implanted dose (see Fig. 12a). The implantation of Au leads to a completely analogous increase of both the spin relaxation time and the ratio $\beta$ (see Fig. 14a and b). Rescaling the implanted densities to take the different defect generation rates for implantation of Ga and Au ions, respectively, into account [29], leads even to a universal dependence on the implanted density (see Fig. 14c and d). This universal dependence strongly suggests that the defects governing the spin dynamics are independent of the implanted ion species.

5.2 Gd-implanted GaN: a room temperature spin injector? Gd-implanted GaN has attracted strong interest as a promising candidate for room temperature spin injectors after experimental reports on ferromagnetism with a Curie temperature far above room temperature [62, 63]. The existence and possible microscopic origins of ferromagnetism in Gd:GaN are, however, controversially discussed both from experimental and theoretical side [64–66]. The ability to efficiently polarize conduction band electrons has also not yet been demonstrated though it is the prerequisite for an application as a spin injector. To shed new light on these questions, we investigated the electron spin dynamics in
Gd-implanted GaN [30]. Different Gd doses corresponding to densities from $0.9 \times 10^{16} \text{ cm}^{-3}$ to $0.9 \times 10^{20} \text{ cm}^{-3}$ were implanted by a focused ion beam implanter into an intentionally undoped wurtzite GaN layer grown on Si(111). The Gd implanted areas were partially coimplanted with Si to investigate the effect of additional defects as well as additional n-type doping. A detailed sample description is given in Ref. [30].

The density dependence of the spin relaxation time $\tau_s$ in Gd-implanted GaN shown in Fig. 15 is in close analogy to the case of implantation of non-magnetic Ga and Au ions as described in the previous section. The spin relaxation time increases with the implantation dose (see Fig. 15a) and, simultaneously, the magnetic field dependence changes from the anisotropic relaxation of free carriers to the $\tau_s \propto 1/B_{\text{ext}}$ dependence typical for localized carriers (cf. Fig. 15b and c). The ratio $\beta = \tau_s / \tau_s$ shows the corresponding transition from $\beta_{\text{gpr}} = 0.75$ to $\beta > 1$ for higher implanted densities (see Fig. 15d). We note that rescaling the implanted densities by the defect generation rates for the different implanted ion species leads to a universal density dependence of the spin relaxation time and the ratio $\beta$ for implantation of Ga, Au, Gd, and Si ions. Therefore, the defect density generated by the implantation governs the electron spin dynamics, independent of the implanted ion species. The observed increase of the spin relaxation time with increasing Gd density is distinctively different from the behavior of Mn-based dilute magnetic semiconductors (DMS), where the spin relaxation time decreases with increasing Mn concentration due to exchange scattering [67]. These findings clearly contradict an efficient magnetic coupling between the implanted Gd ions and the conduction band electrons. As a second step, we investigated the magnetic field dependence of the electronic spin Larmor precession around the external magnetic field, which is a sensitive probe for internal magnetic fields or exchange interactions [70, 71]. The Larmor precession frequency $\omega_L$ depends, however, almost perfectly linear on the external magnetic field in Gd-implanted GaN and shows virtually no dependence on the Gd density (see Fig. 16a and d). These results are in stark contrast to other DMS materials, where a strong renormalization of $\omega_L$ is found [70, 71]. We can therefore rule out an efficient exchange coupling between the conduction band electrons and the Gd ions. The Landé $g$ factor can accordingly be obtained by linear fits to the $\omega_L(B_{\text{ext}})$ dependence. The $g$ factor shows no dependence on the implanted densities and equals the $g$ factor of unimplanted...
GaN. Small effects of an additional splitting $\Delta E_{\text{int}}$ can, however, be probed by the difference $\Delta \omega_h = \omega_h - \omega_{h,\text{GaN}}$ of the Larmor precession frequencies $\omega_h$ of the implanted GaN and $\omega_{h,\text{GaN}}$ of the unimplanted GaN. Though the deviations $\Delta \omega_h$ are very small and do not show a systematic dependence on the implanted density, they are compatible with the values expected for the magnetization $M_{\text{Gd}}$ of comparable samples from literature [68, 69] if one assumes that the magnetization simply adds like a minute contribution to the external magnetic field following $\omega_h = g \mu_B (B_{\text{ext}} + \mu_0 M_{\text{Gd}})/\hbar$.

In conclusion, our results show that the electron spin dynamics in Gd-implanted GaN is governed solely by localization at defects. An efficient exchange coupling between the conduction band electrons and the Gd ions can be ruled out, making an application of Gd-implanted GaN for electron spin injectors highly questionable.

6 Conclusions We have reviewed our experimental results on the electron spin dynamics in bulk GaN by time-resolved magneto-optical Kerr-rotation spectroscopy. The spin relaxation of delocalized electrons is governed by DP relaxation both in the wurtzite and in the cubic phase of GaN. In bulk wurtzite GaN, the symmetry of SOC leads to an intrinsic spin relaxation anisotropy, with spins pointing along the $c$-axis relaxing twice as fast as spins perpendicular to the $c$-axis. The $k$-linear Rashba term completely dominates spin relaxation up to room temperature and up to electron densities of $10^{20} \text{ cm}^{-3}$, and intrinsically limits electron spin lifetimes. In bulk cubic GaN, the $k$-linear Rashba term vanishes due to symmetry and spin relaxation is governed only by the Dresselhaus term, which is weak due to small SOC. Accordingly, we found long spin lifetimes in highly doped cubic GaN that are orders of magnitude larger than in GaAs, making cubic GaN a very promising material system for spintronics.

Spin relaxation in ion-implanted bulk wurtzite GaN is governed by localization at implantation defects, with an increase of spin lifetimes by a factor of 20 for strongly localized electrons. The magnetic field dependence of spin relaxation shows a characteristic change for increasing implantation doses that sensitively probes the degree of localization. Electron spin relaxation in Gd-implanted GaN as a candidate for a room temperature ferromagnetic semiconductor is also completely governed by localization at the implantation defects. An efficient exchange coupling between the conduction band electrons and the magnetic Gd ions can be ruled out, making an application of Gd-implanted GaN for electron spin injectors highly unlikely.

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References

[1] F. Meier and B. P. Zakharchenya (eds.), Optical Orientation (North-Holland, Amsterdam, 1984).
[2] D. D. Awschalom and N. Samarth (eds.), Semiconductor Spintronics and Quantum Computation (Springer-Verlag, Berlin, 2002).
[3] M. I. Dyakonov (ed.), Spin Physics in Semiconductors (Springer-Verlag, Berlin, 2008).
[4] I. Zutic, J. Fabian, and S. D. Sarma, Rev. Mod. Phys. 76, 323–410 (2004).
[5] M. W. Wu, J. H. Jiang, and M. Q. Weng, Phys. Rep. 493, 61–236 (2010).
[6] S. Datta and B. Das, Appl. Phys. Lett. 56, 665–667 (1990).
[7] J. Rudolph, D. Hägele, H. M. Gibbs, G. Khitrova, and M. Oestreicher, Appl. Phys. Lett. 82, 4516–4518 (2003).
[8] J. Rudolph, S. Döhrmann, D. Hägele, M. Oestreicher, and W. Stolz, Appl. Phys. Lett. 87, 241117 (2005).
[9] M. Oestreicher, J. Rudolph, R. Winkler, and D. Hägele, Superlattices Microstruct. 37, 306–312 (2005).
[10] T. Dietl, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, Science 287, 1019 (2000).
[11] S. Nakamura and S. F. Chichibu (eds.), Introduction to Nitride Semiconductor Blue Lasers and Light Emitting Diodes (CRC Press, Boca Raton, 2000).
[12] R. Quay (ed.), Gallium Nitride Electronics (Springer, Berlin, 2008).
[13] D. J. As, in: III-Nitride Semiconductor materials: Growth, lattice Microstruct. 87, 1019 (2000).
[14] L. Lombez, P. F. Braun, H. Carrère, B. Uraszek, P. Renucci, T. Amand, X. Marie, J. C. Harmand, and V. K. Kalevich, Appl. Phys. Lett. 87, 252115 (2005).
[15] D. Lagarde, L. Lombez, X. Marie, A. Balocchi, T. Amand, V. K. Kalevich, A. Shiryaev, E. Ivchenko, and A. Egorov, Phys. Status Solidi A 204, 208–220 (2007).
[16] M. H. Zhao, L. Lombez, B. L. Liu, B. Q. Sun, Q. K. Xue, D. M. Chen, and X. Marie, Appl. Phys. Lett. 95, 041911 (2009).
[17] F. Zhao, A. Balocchi, G. Truong, T. Amand, X. Marie, J. X. Wang, I. A. Buyanova, W. M. Chen, and J. C. Harmand, J. Phys.: Condens. Matter 21, 174211 (2009).
[18] X. Marie, D. Lagarde, V. Kalevich, and T. Amand, in: Dilute III–V Nitride Semiconductors and Material Systems, edited by A. Erol (Springer, Berlin, 2008), chap. 11.
[19] C. Y. Hu, K. Morita, H. Sanada, S. Matsuzaka, Y. Ohno, and H. Ohno, Phys. Rev. B 72, 121203 (2005).
[20] T. Kuroda, T. Yabushita, T. Kosuge, A. Tackeuchi, K. Taniguchi, T. Chinone, and N. Hiro, Appl. Phys. Lett. 85, 3116–3118 (2004).
[21] C. Brimont, M. Gallart, O. Créguet, B. Hönerlage, and P. Gilliot, Phys. Rev. B 77, 125201 (2008).
[22] C. Brimont, M. Gallart, A. Gadalla, O. Créguet, B. Hönerlage, and P. Gilliot, J. Appl. Phys. 105, 023502 (2009).
[23] C. Brimont, M. Gallart, O. Créguet, B. Hönerlage, P. Gilliot, D. Lagarde, A. Balocchi, T. Amand, X. Marie, S. Founta, and H. Mariette, J. Appl. Phys. 106, 053514 (2009).
[24] B. Beschoten, E. Johnston-Halperin, D. K. Young, M. Poggio, J. E. Grimaldi, S. Keller, S. P. DenBaars, U. K. Mishra, E. L. Hu, and D. D. Awschalom, Phys. Rev. B 63, 121202(R) (2001).
[25] J. H. Buß, J. Rudolph, F. Natali, F. Semond, and D. Hägele, Appl. Phys. Lett. 95, 192107 (2009).
