Prediction of extremely long mobile electron spin lifetimes at room temperature in wurtzite semiconductor quantum wells

N. J. Harmon and W. O. Putikka
Department of Physics, Ohio State University, 191 W. Woodruff Ave., Columbus, OH 43210, USA

Robert Joynt
Department of Physics, University of Wisconsin-Madison, 1150 Univ. Ave., Madison, WI 53706, USA

Much of semiconductor research in recent years has focused on the electron spin degree of freedom. For many non-quantum coherent spintronic applications one requires mobile electrons. Even as a carrier of classical information the spin of mobile electrons offers advantages: translation and rotation are in principle dissipationless, offering great potential advantages over charge motion. For all types of spintronic applications, long spin coherence times and ease of spin manipulation at room temperature are of crucial importance. Several types of devices based on mobile spins have been proposed: ballistic [1] and non-ballistic[2] spin field effect transistors, and double-barrier[3] structures. Tuning of the spin-orbit (SO) parameters is possible by applying a gate voltage to a quantum well (QW) to vary the Rashba coupling. In addition, systematic variation of the well width has made it possible to independently tune other couplings and observe momentum-dependent relaxation times in (001)-GaAs QWs.[4]

Experimental studies relevant to the realization of these devices have mainly been carried out at rather low temperatures $T$ or short spin lifetimes $\tau_s$. In GaAs, signs of enhanced lifetime (due to the persistent spin helix[5]) decreased rapidly with temperature and at $T = 300$ K, $\tau_s$ was only slightly above 100 ps; the loss of coherence is due to cubic-in-k terms in the SO Hamiltonian that relax the spin by the D'yakonov-Perel' (DP) mechanism.[6] Theory predicts zinc-blende (zb) (111) QWs benefit at $T = 0$ K from certain cancelations of SO terms.[7] No experiments on this type of QW have been performed. There is experimental evidence that (110)-GaAs QWs have enhanced spin coherence times and theory predicts infinite spin coherence for one element of the relaxation tensor.[8] However, the strong anisotropy of the spin relaxation tensor ensures rapid decoherence in even small magnetic fields.[9] At higher temperatures, long spin lifetimes are limited by the strength of the cubic-in-k SO interaction in direct band gap zb semiconductors. In (110) zb-GaAs, spin relaxation times are limited by intersubband spin relaxation at room temperature.[9]

Nearly all of the theory and experiment along this research direction has been carried out on materials with the zb structure. However, there has also been work on bulk wurtzite (w) materials because their SO splittings are small[10] and there is hope of room temperature ferromagnetism in magnetically doped w-GaN and w-ZnO.[11] Spin dynamics in bulk w-GaN has been studied by two groups,[10] and there has been experimental[12] and theoretical[13, 14] work on spin lifetimes in w-ZnO. This strongly suggests that w-QWs should be studied, and indeed there is some recent work along these lines. Experimentally, the SO splittings have been measured by Lo et al.[15] in Al$_x$Ga$_{1-x}$N/GaN QWs by either Shubnikov-de Haas or weak antilocalization (WAL) measurements. WAL measurements unambiguously point to SO coupling[16] and such measurements are found to agree with theory[17].

Here we determine the general temperature dependence of spin lifetimes in w-QWs due to the DP mechanism. The DP mechanism is dominant at room temperature in bulk zb-GaAs[18] and w-ZnO[13] and this is expected to be true in modulation doped QWs as well. We discuss other relaxation mechanisms that may take over when DP is suppressed and show that they play no role at room temperature. For low-Z w-QWs long spin lifetimes of conduction electrons are possible at low and room temperature due to the combined effects of the w band structure and a small cubic-in-k coefficient in the SO Hamiltonian. We also discuss the feasibility of the needed tunings.

There are striking differences in spin relaxation properties between zb symmetry materials and the hexagonal symmetry of w materials like GaN, ZnO, or AlN. The SO Hamiltonian $H_{SO}$ for (001) QWs with zb symmetry is

$$H_{SO}^b = \alpha_R(k_x \sigma_x - k_y \sigma_y) + \beta_D^b(-k_x \sigma_z + k_y \sigma_y) + \beta_3(k_x k_y^2 \sigma_x - k_y k_x^2 \sigma_y),$$

where $\sigma$ are the Pauli matrices; $\alpha_R$ is the Rashba coupling; $\beta_3$ is the cubic-in-k coupling, while $\beta_3^b = \beta_3 \langle k_z \rangle$.
is a Dresselhaus-type term that is controlled by confinement. \( \langle k_z^2 \rangle \) is the expectation value of the operator \( k_z \) in the QW wavefunction. If \( L \) is the QW width, then \( \langle k_z^2 \rangle \sim (\pi/L)^2 \) for the lowest electric subband and small structural asymmetry. \( \alpha_R \) is proportional to the electric field \( E_z \) and can thus be tuned by applying a gate voltage or producing structures with asymmetry; \( \beta_D^{( zb)} \) depends on \( L \); \( \beta_3 \) depends only on the material and cannot be turned off. When \( \alpha_R = \pm \beta_D^{( zb)} \) the linear-in-\( k \) terms produce a \( k \)-independent effective magnetic field in the \((110)\) or \((\overline{1}10)\) direction.\[2\] This can enhance spin coherence for spins oriented along the effective magnetic field.

For \((001)\) QWs with \( w \) symmetry we have\[15,19\]

\[
H_{SO}^{w} = \left( \alpha_R + \beta_{D}^{(w)} - \beta_3 k_z^2 \right) (ky\sigma_x - k_x\sigma_y),
\]

where \( \beta_{D}^{(w)} = \beta_1 + b\beta_3 \langle k_z^2 \rangle \). This form is clearly different from the \( zb \) case, as has been confirmed experimentally.\[16\] As before, \( \alpha_R \) can be tuned by applying a gate voltage or varying the asymmetry; however, \( \beta_1 \neq 0 \) even in the absence of an applied electric field - the \( w \) structure does not have the mirror symmetry \( z \leftrightarrow -z \). \( \beta_{D}^{(w)} \) can be tuned by changing \( L \).

There are two important formal differences between \( H_{SO}^{zb} \) and \( H_{SO}^{w} \). First, at the linear-in-\( k \) level, setting \( \alpha_R = -\beta_{D}^{(w)} \) in \( H_{SO}^{w} \) gives zero effective magnetic field for all \( k \), which means much more dramatic enhancement of spin coherence. As pointed out by Lo et al., this is what makes their \( Al_xGa_{1-x}N/GaN \) structure an excellent candidate for the non-ballistic spin field effect transistor.\[15\] Second, if we define \( k_\parallel = (k_x, k_y) \), then at the cubic-in-\( k \) level for a circular Fermi surface and elastic scattering, \( |k_\parallel| \) is conserved and we can set \( k_\parallel = k_F \), the Fermi wavevector. As pointed out by Wang et al., the effective field can be cancelled \( all \) the way to third order by enforcing the condition \( \alpha_R + \beta_{D}^{(w)} - \beta_3 k_z^2 = 0 \), eliminating what appears to be the major source of spin decoherence in the experiments to date.\[15,19\] Note that the final term can be independently tuned by changing the electron density. The SO Hamiltonian for \((111)\) \( zb \)-QWs is similar to \((001)\) \( w \)-QWs;\[27\] similarities also exist between \((110)\) \( zb \) and \((100)/(010)\) \( w \)-QWs which will be dealt with in a future publication.

In addition to different crystal symmetries, an important quantitative difference between \( zb \) and \( w \) semiconductors is the strength of the SO interaction. The common wurtzite GaN has smaller SO coupling than GaAs. Of prime importance for spin lifetimes at room temperature is the coefficient for the cubic-in-\( k \) terms in the SO Hamiltonian: \( \beta_3 = -0.32 \) meV nm\(^3\) for GaN\[22\] compared to \( \beta_3 = 6.5 - 30 \) meV nm\(^3\) for GaAs.\[4\] From the spin lifetimes evaluated below, an expression for the maximum \( \tau_s \) at high temperatures, \( T \gg T_F \), can be obtained analytically \( \tau_s^{-1}(\max) = 32\tau_{tr}m^3\beta_3^2k_B^3T^3/h^8 \), which shows that \( \beta_3 \) determines the fall-off of \( \tau_s(\max) \). w-AlN is the most favorable, with \( \beta_3 = -0.01 \) meV nm\(^3\), but is less well-characterized than GaN since experiments are lacking and only one theoretical estimate of \( \beta_1 \) has been calculated.\[19,21,23\]

In Fig. 1(a) we plot \( \tau_s \) as a function of \( \alpha_R \) at \( T = 300 \) K for w-AlN and w-GaN, with the aforementioned parameter values. The SO couplings of w-GaN are the best-characterized of the \( w \) materials. When \( \alpha_R \) is appropriately tuned for w-GaN, \( \tau_s \) approaches \( 10 \) \( \mu s \) at \( T = 5 \) K if \( \tau_{tr} \approx 0.1 \) ps and even at \( T = 300 \) K can reach values of \( 4 \text{ ns} \) if \( \tau_{tr} \approx 0.1 \) ps. At \( T = 300 \) K and \( \tau_{tr} \approx 0.1 \) ps, the maximum spin lifetime in w-AlN is \( 0.5 \) \( \mu s \). At \( 5 \) K, the spin lifetime in w-AlN surpasses \( 2 \) ms if \( \tau_{tr} \approx 1 \) ps. The peaks in \( \tau_s \) lie slightly off the condition \( \alpha_R = -\beta_D^{(w)} \); the difference is a measure of the importance of the cubic-in-\( k \) term. This implies that devices meant to be operated at different temperatures would need to be tuned somewhat differently. The extremely long spin relaxation times in both w-GaN and w-AlN are not limited by the Elliott-Yafet (EY) mechanism\[24\] since we determine the spin relaxation time due to EY to be \( \tau_{EY} \sim 100\mu s \) at room temperature. Unlike in \((110) \) \( zb \)-QWs, intersubband spin relaxation is not the limiting mechanism either; due to the small SO coupling we find it to be \( 1 \text{ ms} - 10^6 \) times weaker than in zb-GaAs QWs.\[9\] As a comparison to the \( w \) materials we show the corresponding high temperature calculations for \((001)\) and \((111) \) \( zb \)-GaAs QWs in Fig. 1(b). The times are shorter by orders of magnitude compared to the \( w \) materials. High temperature maxima expressions for zb-(001) and zb-(111) are similar to what was determined for w-(001). The contrast in \( \tau_s \) is due to the much larger \( \beta_3 \) in GaAs.

We now sketch the calculation that gives the results quoted above. The spin relaxation tensor \( \Gamma_{ij} \) is defined by the equation \( dS_i/dt = -\sum_{j=x,y,z} \Gamma_{ij} S_j \) and \( S \) is the spin polarization vector. The DP contribution to \( \Gamma \) is given by \[20\]

\[
\Gamma_{ij}(T) = \frac{1}{2\hbar^2} \sum_{n=-\infty}^{\infty} \int_{-\infty}^{\infty} dE f_+(E,T) - f_-(E,T) |\tau_n\gamma_{ij}^{(n)}|^2 \int_{-\infty}^{\infty} dE f_+(E,T) - f_-(E,T),
\]

where \( \gamma_{ij}^{(n)}(k_\parallel) = \text{Tr} \{ H_{SO}^{(n)} [H_{SO}^{(n)},\sigma_j] \sigma_i \} \). The scattering rates are given by \( 1/\tau_n = \int_{0}^{2\pi} d\phi_k \text{Tr} \{ H_{SO}^{(n)}(k_\parallel) \exp(-i\phi_k) \} \), \( \phi_k \) is the angle between \( k_\parallel \) and the \( k_x \)-axis, \( \theta \) is the scattering angle, and \( W(E,\theta) \) is the scattering rate. The various scattering times are energy-dependent due to the energy dependence of \( W \). Here we take \( \tau_1 \) to be energy independent for simplicity and to illustrate the main ideas. We reserve a full treatment of the energy dependence for future work.

We focus first on the \( w \) case with a circular Fermi surface. When the energy splitting between positive and
negative helicity states is small, we find
\[ \Gamma'(\omega) = \frac{1}{2} \Gamma'(\omega) = \frac{2\tau_F}{\hbar^2 I_0(\beta\mu)} \left[ (\alpha_R + \beta_D(\omega))^2 \zeta I_3(\beta\mu) \right] 
- 2(\alpha_R + \beta_D(\omega))\beta\zeta \frac{\zeta I_2(\beta\mu) + \beta_D^2\zeta I_3(\beta\mu)}{\beta} \right], \tag{1} \]
where \( \zeta = 2m^*k_B T/\hbar^2 \), \( \beta = 1/k_B T \), \( \mu \) is the chemical potential, \( I_r(z) \equiv \int_0^\infty dx x^r / \left( 4 \cosh^2 (x - z) / 2 \right) \), and \( \tau_F \), the transport time, is an experimental quantity. All other components of \( \Gamma \) vanish. This simplifies at zero temperature \( \Gamma'(\omega) = 4\tau_m E_F(\alpha_R + \beta_D(\omega) - 2m^*\beta_D E_F h^{-2} h^2 \). \( E_F \) is the Fermi energy. Clearly the \( T = 0 \) relaxation times diverge when the tunable quantity \( \alpha_R + \beta_D(\omega) - 2m^*\beta_D E_F h^{-2} h^2 \) vanishes. This divergence is cut off by finite temperatures.

In zb structures \( \Gamma \) is observed to be anisotropic, and the appropriate quantities are \( \Gamma^{(zb)}_+ \) and \( \Gamma^{(zb)}_z \) the relaxation rates for spin along the [110] (||110||) and [001] directions. The results are determined in Ref. [20]. These expressions for zb approach finite limits as \( T \to 0 \), regardless of the values of the parameters. This is in sharp contrast to the relaxation rates in Eq. [1].

We now address the tunability of possible devices. In zb-GaAs it has been possible to achieve quite substantial variations in the appropriate parameters. [4, 25] Koralek et al. were able to change \( \beta^{(zb)}_D \) by making structures with different well widths and to change \( \alpha_R \) by adjusting the dopant concentrations on the sides of the well, corresponding to a maximum electric field of \( 5.4 \times 10^{-3} \) V/nm. In this way a range of \( \alpha_R/\beta^{(zb)}_D \) of about 0.25 to 1.25 was achieved, without even needing a gate. \( \beta_3 \) was inaccessible and remained constant for all structures, setting an upper limit on spin lifetimes. Significant experimental tuning of SO coupling in w-GaN has not yet been achieved. However, calculations have been done [26] for w-GaN, which produce the correct magnitude for the spin splittings overall (\( \sim 5 \) meV at Fermi wavevectors of typical structures).

These authors do not compute \( \alpha_R, \beta_1, \) and \( \beta_3 \) explicitly, but their computed spin splittings at a typical Fermi wavevector shows that changes in spin splittings by a factor of 4 or so can be achieved by changing the well width from 10 to 2 unit cells; to achieve the same sort of change due to external electric fields required very strong fields of order 1 V/nm. This suggests that changing well width and electron density rather than electric field will be the most favorable route for tuning of w-QWs.

In conclusion, we predict that QWs consisting of w materials, e.g. GaN/GaAlN, can be tuned to achieve very long spin lifetimes. These lifetimes are at zero momentum (\( \mathbf{q} = 0 \)), not helical modes, and are therefore better for spin injection and transistor devices operating at mesoscopic length scales. We predict AlN may offer the longest spin lifetimes, with a lifetime of 0.5 \( \mu \)s at room temperature.

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[1] S. Datta and B. Das, Appl. Phys. Lett. 56, 665 (1990).
[2] J. Schliemann, J. C. Egues, and D. Loss, Phys. Rev. Lett. 90, 146801 (2003); X. Cartoixa, D.Z.Y. Ting, and Y.C. Chang, Appl. Phys. Lett. 83, 1462 (2003).
[3] K.C. Hall and M.E. Flatté, Appl. Phys. Lett. 88, 162503 (2006).
[4] J.D. Koralek, C.P. Weber, J. Orenstein, B.A. Bernevig, S.-C. Zhang, S. Mack, and D.D. Awschalom, Nature 458, 610 (2009).
[5] B.A. Bernevig, J.O. Orenstein, and X.-C. Zhang, Phys. Rev. Lett. 97, 236601 (2006).
[6] M. I. Dyakonov and V. I. Perel, Sov. Phys. JETP 33, 1053 (1971).
[7] X. Cartoixa, D.Z.Y. Ting, and Y.C. Chang, Phys. Rev. B 71, 045313 (2005).
[8] M. I. Dyakonov and V. Y. Kachorovskii, Sov. Phys. Semicond. 20, 110 (1986); Y. Ohno, R. Terauchi, T. Adachi, F. Matsukura, and H. Ohno, Phys. Rev. Lett. 83, 4196 (1999).
[9] W.J. Doehrmann, J.B. Hagele, P.A. Rudolph, S.T. Bich-
[10] 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); J.H. Buß, J. Rudolph, F. Natali, F. Semond, and D. Hagele, Appl. Phys. Lett. 95, 192107 (2009).

[11] C. Liu, F. Yun, and H. Morkoc, J. Mater. Sci.: Mater. Electron. 16, 555 (2005).

[12] S. Ghosh, V. Sih, W.H. Lau, and D.D. Awschalom, Appl. Phys. Lett. 86, 232507 (2005); S. Ghosh, D.W. Steuerman, B. Maertz, K. Ohtani, Huaizhe Xu, H. Ohno, and D.D. Awschalom, Appl. Phys. Lett. 92, 162109 (2008).

[13] N.J. Harmon, W.O. Putikka, and R. Joynt, Phys. Rev. B 79, 115204 (2009).

[14] C. Lü and J.L. Cheng, Semicond. Sci. Technol. 24, 115010 (2009).

[15] I. Lo, M.H. Gau, J.K. Tsai, Y.L. Chen, Z.J. Chang, W.T. Wang, J.C. Chiang, and T. Aggerstam, S. Lourdudoss, Phys. Rev. B 75, 245307 (2007).

[16] N. Thillosen, S. Cabañas, N. Kaluza, V.A. Guzenko, H. Hardtdegen, and Th. Schäpers, Phys. Rev. B 73, 241311(R) (2006).

[17] S.B. Lisesivdin, N. Balkan, O. Makarovský, A. Patane, A. Yıldız, M.D. Caliskan, M. Kasap, S. Ozcelik, and E. Ozbay, J. Appl. Phys. 105, 093701 (2009).

[18] W.O. Putikka and R. Joynt, Phys. Rev. B 70, 113201 (2004).

[19] Wan-Tsang Wang, C.L. Wu, S.F. Tsay, M.H. Gau, Ibai Lo, H.F. Kao, D.J. Jang, Jih-Chen Chiang, Meng-En Lee, Yia-Chung Chang, Chun-Nan Chen, and H.C. Hsueh, Appl. Phys. Lett. 91, 082110 (2007).

[20] J. Kainz, U. Rössler and R. Winkler, Phys. Rev. B 70, 195322 (2004).

[21] J. Nicklas, unpublished density functional theory calculation.

[22] J.Y. Fu and M.W. Wu, J. Appl. Phys. 104, 093712 (2008).

[23] private communication with Meng-En Lee, author of Ref. [19].

[24] R.J. Elliott, Phys. Rev. 96, 266 (1954); A. Tackeuchi, T. Kuroda, S. Muto, Y. Nishikawa, and O. Wada, Jpn. J. Appl. Phys. 38, 4680 (1999).

[25] M. Studer, G. Salis, K. Ensslin, D.C. Driscoll, and A.C. Gossard, Phys. Rev. Lett. 103, 027201 (2009).

[26] I. Lo, W.T. Wang, M.H. Gau, J.K. Tsai, S.F. Tsay, and J.C. Chiang, Appl. Phys. Lett. 88, 082108 (2006).