Experimental determination of Rashba spin-orbit coupling in wurtzite \( n \)-GaN:Si

W. Stefanowicz,¹ R. Adhikari,² T. Andraezyk,¹ B. Faina,² M. Sawicki,¹ J. A. Majewski,³ T. Dietl,¹,3,4 and A. Bonanni²

¹Institute of Physics, Polish Academy of Sciences, al. Lotników 32/46, PL-02 668 Warszawa, Poland
²Institut für Halbleiter-und-Festkörperphysik, Johannes Kepler University, Altenbergerstr. 69, A-4040 Linz, Austria
³Institute of Theoretical Physics, Faculty of Physics, University of Warsaw, ul. Hoża 69, PL-00 681 Warszawa, Poland
⁴WPI-Advanced Institute for Materials Research (WPI-AIMR), Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai 980-8577, Japan

(Dated: March 3, 2014)

Millikelvin magnetotransport studies are carried out on heavily \( n \)-doped wurtzite GaN:Si films grown on semi-insulating GaN:Mn buffer layers by metal-organic vapor phase epitaxy. The dependence of the conductivity on magnetic field and temperature is interpreted in terms of theories that take into account disorder-induced quantum interference of one-electron and many-electron self-crossing trajectories. The Rashba parameter \( \alpha_R = (4.5 \pm 1) \text{meVÅ} \) is determined, and it is shown that in the previous studies of electrons adjacent to GaN/(Al,Ga)N interfaces, bulk inversion asymmetry was dominant over structural inversion asymmetry. The comparison of experimental and theoretical values of \( \alpha_R \) across a series of wurtzite semiconductors is presented as a test of current relativistic \textit{ab initio} computation schemes. It is found that electron-electron scattering with small energy transfer accounts for low temperature decoherence in these systems.

PACS numbers: 71.70.Ej, 72.15.Rn, 72.80.Ey, 72.20.-i

Beside being strategic materials systems for nowadays optoelectronic and high-power applications GaN and related alloys are expected to play a major role in the realization of spin-related functionalities based on semiconductors. In particular, Fe doping serves routinely to fabricate semi-insulating GaN substrates. However, GaN with higher Fe concentrations exhibits room temperature ferromagnetic and antiferromagnetic features associated with the aggregation of Fe cations, leading to the formation of various magnetically robust Fe\(_2\)N nanocrystals. Furthermore, light absorption associated with the Mn mid-gap band in GaN improves the efficiency of GaN-based solar cells. At the same time, the formation of Mn-Mg\(_x\) impurity complexes activates room temperature infrared luminescence suggesting that the optoelectronic capabilities of nitrides can be extended towards the communication windows.

Other appealing aspects of these systems are associated with spin-orbit coupling (SOC). On one hand, the small value of the nitrogen proton number \( Z_n \), leading to weak spin-orbit splitting \( \Delta_{so} \) of the valence band, results in a long spin relaxation time of electrons in GaN. On the other hand, strong interfacial electric fields result in substantial Rashba-type SOC that in the extreme case of GaN/InN/GaN quantum wells may lead to a transition to the topological insulator phase. In this context particularly appealing is the demonstration that (Ga,Mn)N is a ferromagnetic insulator which offers prospects to the search for phenomena associated with the interplay between SOC and the exchange splitting of bands by ferromagnetic proximity effects.

Here, we report on experimental studies of weak localization and antilocalization magnetoresistance in epitaxial layers of wurtzite (wz) \( n \)-GaN:Si. A theoretical description of the data in terms of the Hikami, Larkin, and Nagaoka theory is suitably adopted for wz compounds and allows us to extract the magnitude of the parameter \( \alpha_R \) describing the Rashba term linear in \( k \) and accounting for the spin-splitting of the conduction band in bulk wz semiconductors. The value of \( \alpha_R = (4.5 \pm 1) \text{meVÅ} \) we determine here is by two orders of magnitude greater than the one found by electron spin resonance (ESR) for electrons trapped by donors or accumulated at the surface of \( n \)-GaN. At the same time, it agrees with the value found for electrons attracted to the GaN/(Al,Ga)N interface by polarization electric fields. Our results demonstrate, therefore, that the bulk rather than the structure inversion asymmetry accounts for the spin splitting of interfacial states. We discuss also the chemical trends in \( \alpha_R \) and show that the discrepancy between the current \textit{ab initio} theories and the present and previous magnetoresistance\( \text{ and ESR} \) studies is within a factor of two for various wz-\( n \)-type semiconductors: ZnO, GaN, CdS, and CdSe. Finally, we treat the temperature dependence of the phase coherence length \( L_{\phi} \) and conductivity \( \sigma \) in terms of electron-electron interactions in disordered systems.

The Si-doped GaN layers considered in the present study have been grown in an AIXTRON 200RF horizontal tube metalorganic vapor phase epitaxy (MOVPE) reactor and deposited on a c-plane sapphire substrate using TMG\(_x\), MnC\(_{2p}\), NH\(_3\), and SiH\(_4\) as precursors for Ga, Mn, N, and Si respectively, with H\(_2\) as carrier gas. After nitridation of the sapphire substrate, a low temperature nucleation layer (NL) is deposited at 540°C and then annealed at 1040°C. Successively, a 1 μm-thick GaN:Mn
buffer layer is grown also at 1040°C, Mn being introduced in order to compensate the n-type background proper of the GaN layers fabricated by MOVPE. The concentration of Mn in the buffer layer is as low as 0.06%, as confirmed by secondary-ion mass spectroscopy (SIMS) and SQUID magnetometry. A 150 nm layer of GaN:Si is further grown at 1000°C onto the GaN:Mn buffer layer. All steps of the growth process are monitored with in situ spectroscopic and kinetic ellipsometry.

The grown samples are systematically characterized by atomic force microscope (AFM), high resolution x-ray diffraction (HRXRD), high resolution transmission electron microscopy (HRTEM) and SIMS has been employed for chemical analysis. The AFM micrographs reveal a flat surface (rms roughness $\approx 1$ nm) while HRXRD and HRTEM confirm the high crystallinity of the samples. The HRTEM analysis does not reveal any secondary phases like e.g., precipitates of Si$_3$N and energy dispersive x-ray spectroscopy (EDS) states the homogeneous distribution of Si in the doped layer with a concentration $\approx 7\times 10^{19}$ cm$^{-3}$, far over the critical value for the metal-to-insulator (MIT) transition in bulk GaN, $n_{MIT} \approx 10^{18}$ cm$^{-3}$ (Ref. 33). The degenerate and metallic character of the samples is further documented by the absence of dependence of the $\mu$, resistivity, $\rho$, and $k_F\ell$ as a function of the magnetic field in the limit $T \to 0$, as well as by the values of the Hall mobility $\mu = 140$ cm$^2$/V s and $k_F\ell = 4.6$ in this regime, where $\ell = h\k_F\mu/e$ is the mean free path. Accordingly, we interpret the measured magnetoresistance $\Delta\rho(T, B)$ in terms of quantum corrections to the conductivity of disordered systems, developed for $k_F\ell > 1$ and $\ell < l_B = (h/eB)^{1/2}$ (Refs. 32 and 33).

In Figs. 1 and 2 the conductivity $\sigma(T, B) = 1/\rho(T, B)$ of the GaN:Si film is shown at different temperatures $T$ as a function of the magnetic field $B$ applied perpendicular to the film surface, i.e., parallel to the wz-c-axis. It is seen from Fig. 1 that for $T \geq 10$ K the magnetoconductivity (MC) is solely positive, while from the data collected in Fig. 2 for $T \leq 1.5$ K and down to 40 mK there are contributions of both negative and positive MC in low magnetic fields. This negative component of MC is related to the appearance of a weak antilocalization (WAL) maximum (which vanishes above 1 K), a distinct signature of SOC.

For $T \geq 10$ K, the experimental results are fitted within a three dimensional (3D) theoretical model of weak localization MC in semiconductors on the metallic side of the MIT, as proposed by Kawabata (points: experimental data; solid lines: theoretical fitting). The 3D theory of conductivity changes in the magnetic field by Kawabata is employed with the phase coherence length $L_c(T)$ as the only fitting parameter. Here, the theory describes the data quite accurately. We add that the presence of positive MC is commonly taken as an indication for spin disorder scattering. Actually, even in the presence of magnetic impurities, spin disorder scattering is typically masked by other scattering mechanisms in semiconductors, and rarely perturbs the conductivity directly.

However, this 3D model does not describe the observed MC for $T \leq 5$ K and low magnetic fields, where two additional aspects must be considered, namely: (i) the impact of SOC on the quantum corrections to the conductivity, leading to a WAL maximum in MC below 1 K (as seen in Fig. 2) and (ii) a dimensional crossover from 3D to 2D that occurs if $L_c(T) \gtrsim d$, where $d$ is the layer thickness. In Fig. 2 the fingerprint of WAL is observed for $T \leq 1$ K and for fields $\lesssim 1$ mT. The MC data obtained for $T \leq 1.5$ K are fitted with the theoretical model proposed for 2D films in the weakly localized regime, $k_F\ell > 1$, and considering effects of spin-dependent scattering.

According to the $\bm{k} \cdot \bm{p}$ theory, SOC in wz semiconductors leads to a term linear in $k$ in the effective mass equation:

$$
H_{so} = \alpha_R \hat{c} \cdot (\vec{\sigma} \times \vec{k}),
$$

where $\alpha_R$ is the Rashba $\bm{k} \cdot \bm{p}$ parameter; $\hat{c}$ is the versor along the wz c-axis, and $\vec{\sigma}$ are the Pauli matrices. For $\alpha_R k_F\tau/\hbar < 1$, the corresponding spin relaxation times are given by:

$$
\tau_{so}^{-1} = \tau_{so}^{-1} = \alpha_R^2 k_F^2 \tau/3\hbar^2; \tau_{so}^{-1} = 0,
$$

where the $z$ axis is taken along the wz c-axis, $\tau = \mu m^*/e$ is the momentum relaxation time, and effective mass is $m^* = 0.22m_0$ for GaN (Ref. 40).

Here, two fitting parameters $\alpha_R$ and $L_c(T)$, are employed to describe the conductivity changes in mag-
netic field. From the fitting of the MC data in the low temperature range, we find \( \alpha_R \) for wz \( n \)-GaN:Si to be \((4.5 \pm 1) \text{ meV} \alpha \). Within the experimental uncertainties, this value is virtually identical to the one determined non-perturbatively from various experimental methods for wz \( n \)-GaN:Si to be \((4.5 \pm 1) \text{ meV} \alpha \). Within the experimental uncertainties, this value is virtually identical to the one determined through the interfacial field by Hikami et al., treating \( \alpha_R \) and \( L_\varphi(T) \) as fitting parameters. The two lowest curves are down-shifted for clarity.

In Fig. 2 we provide a compilation of \( \alpha_R \) values determined through various experimental methods for wz semiconductor compounds, plotted as a function of a harmonic average of the cation and anion proton numbers, \( \bar{Z} = 2/(1/Z_c + 1/Z_a)^{-1} \), and compared to results of \( ab \) initio computations in the framework of the density functional theory with relativistic effects taken into account non-perturbatively. A chemical trend, \( \alpha_R \approx \bar{Z}^{2.2 \pm 0.5} \), is evident and confirms that the significance of the SOC increases with the nucleus charge. Furthermore, it is seen that the theory describes the experimental values with an accuracy better than a factor of two.

Our fitting procedure provides also the values of \( L_\varphi(T) \), which are shown in Fig. 3 and compared to corresponding data for CdSe:In (Ref. 16) and ZnO:Al (Ref. 28). A dependence \( L_\varphi(T) = aT^{-3/4} \) is observed for all compounds over a wide temperature range, a behavior expected theoretically for decoherence brought about by electron-electron interactions in 3D (Refs. 32 and 33). A transition to the 2D case occurs in this case at \( L_T = \hbar(k_F^2/3k_B T m^*)^{1/2} \approx d \). A change in the \( L_\varphi(T) \) slope observed in this region, if not caused by noise-related decoherence, can be associated with the dimensional cross-over.

According to theoretical expectations, \( L_\varphi(T) \) has the same functional form for electron-electron scattering with large and small energy transfers in the 3D case. Our quantitative evaluation demonstrates that low energy processes dominate. They correspond to decoherence by electromagnetic fields generated by thermal fluctuations of the electron liquid, for which

\[
L_\varphi(T) = k_F(\ell L_T^3/3\pi^3)^{1/2}.
\]

According to this theory, the prefactor \( a \) is \( 214, 224, \) and \( 964 \text{ nmK}^{3/4} \) for samples of CdSe:In, GaN:Si, and ZnO:Al, respectively. These values are in good agreement with experimental data corresponding to the results in Fig. 3, namely \( a \approx 300, 440, \) and \( 1200 \text{ nmK}^{3/4} \).

In Fig. 5 the zero magnetic field conductivity \( \sigma(T) \) is reported as a function of \( T^{0.5} \), and it is seen to have a linear dependence on square root temperature below 25 K. This behavior is assigned to quantum corrections to the conductivity due to disorder-modified electron-electron interactions. A quantitative comparison of this slope to the theory leads to a value of the coupling parameter in the triplet channel \( F \approx 0.14 \). This relatively small...
value can be expected for $k_B \ell \gg 1$. However, on approaching the metal-to-insulator transition the value of $F$ increases. This effect was observed in n-CdSe, where $F \approx 2.7$ and the conductivity decreases with increasing temperature.\textsuperscript{16}

In summary, we have carried out low temperature magnetotransport studies on high quality heavily doped wz-GaN:Si films grown on a semi-insulating GaN:Mn buffer layer. Our investigations reconfirm the relevance, in doped degenerate semiconductors, of disorder-induced quantum interference of one-electron and many-electron self-crossing trajectories, effects not captured by the Drude-Boltzmann description of transport phenomena. The quantitative models of the magnetocconductance data have allowed us to determine the Rashba parameter $\alpha_R$, so far known only from studies of 2DEG adjacent to GaN/(Al,Ga)N interfaces. Our results demonstrate that inversion asymmetry associated with the wurtzite crystal structure dominates over the effects of the interfacial electric field in the conduction band of GaN. The comparison of experimental and theoretical values of $\alpha_R$ across a series of wz semiconductors has provided an important test of the current relativistic ab initio computation schemes, demonstrating that the differences between the experimental and theoretical values are within a factor of two. Furthermore, our quantitative interpretation of the decoherence length $L_\varphi(T)$ has shown that electromagnetic fields brought about by thermal fluctuations of electron liquid account for the low temperature decoherence of conducting electrons in these systems. With these premises, wide perspectives open for nitrides, as building-blocks for the next generation of spin devices exploiting spin-orbit coupling and the magnetism of transition-metal doped layers.

This work was supported by the FundMS Advanced Grant of the European Research Council (ERC Grant No. 227690) within the Ideas 7th Framework Programme of the European Community, and by the Austrian Science Foundation – FWF (P20065, P22477, and P24471), and by Polish National Science Centre under grant No. 2011/03/B/ST3/02457.
