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Spin precession of holes in wurtzite GaN studied using the time-resolved Kerr rotation technique

C. Y. Hu,[1,2,*] K. Morita,[1,3] H. Sanada,[1] S. Matsuzaka,[1] Y. Ohno,[1,2,†] and H. Ohno[1,3,‡]
[1]*Laboratory for Nanoelectronics and Spintronics, Research Institute of Electrical Communication, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan*
[2]*CREST, Japan Science and Technology Agency, Japan*
[3]*ERATO Semiconductor Spintronics Project, Japan Science and Technology Agency, Japan*

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The coherent spin dynamics of holes in Mg-doped (*p*-type) wurtzite GaN is studied by time-resolved Kerr rotation technique. We observe the spin precession of holes with the spin coherence time of 120 ps, which is distinguished from that of electrons by comparison with the results in Si-doped *n*-GaN. The *g* factor anisotropy of holes is determined to be $g^h_\perp = 2.17 \pm 0.03$ and $g^h_\parallel = 2.27 \pm 0.03$. We identify that the involved holes are in the valence band *B* (upper Γ_7) of wurtzite GaN.

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Wide gap III-V semiconductor GaN is one of fascinating materials for short wavelength optoelectronic devices. In addition, GaN is one of the candidates for achieving room-temperature ferromagnetism in semiconductors.[1] Thus, it is of great importance to understand the spin dynamics of carriers and excitons in GaN for possible applications to spintronics devices and spin-based quantum information processing.[2] Compared to other major III-V and II-VI semiconductors such as GaAs (Refs. 3 and 4) and ZnSe,[5] however, there exists only few experimental reports of coherent spin dynamics[6] and relaxation processes of conduction electrons[7] in GaN. For GaN, to our best knowledge, there is no report on the spin dynamics of holes, which is rarely reported even in GaAs-based structures due to the short spin coherence time as a result of the strong valence-band mixing.[8]

In wurtzite GaN, the valence-bands at the Brillouin zone center are split into doubly degenerate Γ_9 (labeled as *A*), upper Γ_7 (*B*), and lower Γ_7 (*C*) bands by crystal-field and spin-orbit effects. A fundamental band parameter, the *g* factor anisotropy of the valence band is still unknown.[9] For *p*-type doping, Mg is a promising acceptor in GaN to achieve high density of holes, and it should have strong anisotropic g factor in the effective mass approximation. On the other hand, electron paramagnetic resonance (EPR) and optically detected magnetic resonance (ODMR) experiments give nearly isotropic *g* factor of the Mg acceptor: $g_\parallel = 2.1$, $g_\perp = 2.0$, and $\Delta g = 0.1$.[10,11] The understanding of this puzzle is attracting both theoretical and experimental investigations.[12,13]

In this paper, we report the observation of spin precession of holes in wurtzite Mg-doped (*p*-type) GaN by using time-resolved Kerr rotation (TRKR) technique. This enables us to determine the spin coherence time and *g* factor anisotropy of holes in wurtzite GaN through the observation of coherent spin precession in transverse magnetic fields. As a reference, we also measure the spin precession of electrons in conduction band in Si-doped *n*-type GaN.

The samples studied in this paper are 2-μm-thick GaN epilayers on (0001) sapphire substrates.[14] The *n*-type GaN epilayer is Si-doped with an electron density of $\sim 1 \times 10^{18}$ cm^{-3}, and the *p*-type GaN epilayers are Mg-doped with a hole density of $\sim 5 \times 10^{17}$ cm^{-3} (two pieces #1 and #2 are cut from the same wafer). Between the substrate and the epilayer is a 2-μm-thick undoped GaN buffer layer.

The samples were placed in an optical cryostat with a split-coil magnetic field system (temperature 1.7–300 K and magnetic field 0–7 T). The angle α between the magnetic field and *c*-axis (i.e., growth direction) can be varied by rotating the sample rod. TRKR and pump-probe experiments are performed by using a mode-locked Ti:sapphire laser with a repetition rate of 76 MHz and a regenerative amplifier (RegA) operating at 100 kHz. The RegA is seeded by the mode-locked 100 fs laser pulse centered at 800 nm. The output of the RegA was used to drive an optical parametric amplifier with the generated signal beam tunable from 500 nm to 740 nm. The signal beam was then frequency-doubled by a second-harmonic generator to the wavelength range 250–370 nm (or photon energy of 3.35–4.96 eV). The final laser pulses are split into the pump beam and probe beam. The pump beam is circularly polarized (in order to excite spins of carriers) and the power is kept at 1.6 mW. The probe beam is linearly polarized with a power of 0.3 mW. Both beams are focused onto the sample with a spot size of ~ 100 μm. The delay time between the pump and the probe beam can be scanned from 0 to 600 ps. The Kerr rotation angle of the probe beam reflected from the sample is analyzed by an optical bridge made of two UV-enhanced Si photodiodes. Similar to time-resolved Faraday rotation (TRFR) (but different mechanisms), TRKR signals are proportional to the spin component along the propagation direction of the probe beam, and TRKR signals versus the delay time between the pump and probe beam map the coherent spin evolution or spin precession of carriers if a transverse magnetic field is applied.

Figure 1 shows the TRKR curves taken from *n*-type GaN at 10 K. When the pump beam changes from right-circular (σ^+) to left-circular (σ^-) polarizations or vice versa, the phase of TRKR curves changes π as the direction of excited spins is reversed [see Fig. 1(a)]. So we just present TRKR curves with pump beam σ^+ polarized in Figs. 1(b) and 1(c). The observed oscillations originate from spin precession of

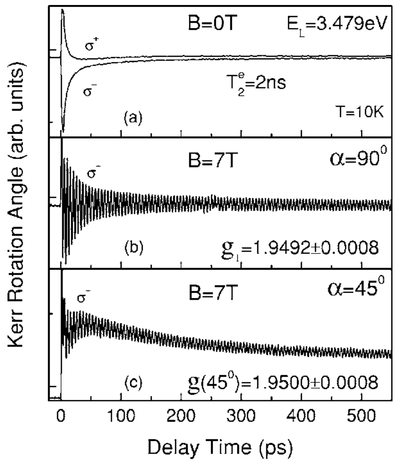

FIG. 1. Time-resolved Kerr rotation (TRKR) taken from an *n*-type GaN epilayer at 10 K and (a) $B=0$; (b) $B=7$ T at $\alpha=90°$; (c) $B=7$ T at $\alpha=45°$. The laser energy is $E_L=3.479$ eV and the pump beam is either σ^+ or σ^- polarized.

carriers under magnetic fields. From the oscillation period T, the effective g factor can be deduced from $h/T=g(\alpha)\mu_B B$, where h is the Planck constant and μ_B is the Bohr magneton. We get $g(90°)=1.9492\pm0.0008$ and $g(45°)=1.9500\pm0.0008$. $g(\alpha)$ can be expressed by

$$g(\alpha) = \sqrt{g_{\parallel}^2 \cos^2 \alpha + g_{\perp}^2 \sin^2 \alpha}, \quad (1)$$

where $g_{\parallel}$ and $g_{\perp}$ are longitudinal and transverse g factor with magnetic field along and perpendicular to c-axis respectively. We can determine $g_{\perp}=1.9492\pm0.0008$ and $g_{\parallel}=1.9508\pm0.0008$. These values of high precision are the same as the g factor values of electrons in GaN measured by ESR experiments.[15] Therefore, we identify that the observed spin precession comes from electrons in the conduction band.

By checking the curves in Fig. 1(a), we find there are three slopes with the corresponding decay time constants of 15 ps, 160 ps, and 2 ns.[16] This phenomenon is similar to the TRFR results in *n*-GaN reported by Beschoten *et al.*[6] The two shorter decay times are related to the spin relaxation and recombination of holes which induce the decoherence to electron spins, and the longer decay time corresponds to the spin coherence time of electrons, i.e., $T_2^{e*}=2$ ns, in accordance with Ref. 6.

After identifying the spin precession of electrons in *n*-type GaN, we now turn to study *p*-type GaN by TRKR. Figure 2 presents the pump-probe measurements in *p*-type GaN at 10 K. The electron-hole recombination time is measured to be 1.3 ns. This lifetime does not change when the laser energy is tuned from 3.446 eV to 3.550 eV around the GaN band gap (∼3.47 eV).

Figure 3 presents the TRKR curves taken from *p*-type GaN at 10 K with the laser energy tuned at 3.482 eV. Following the same procedure as for electrons in *n*-GaN, we obtain $g(90°)=2.17\pm0.03$ and $g(45°)=2.22\pm0.03$, or $g_{\perp}=2.17\pm0.03$ and $g_{\parallel}=2.27\pm0.03$, which are different from the g factor values of electrons in the conduction band. So

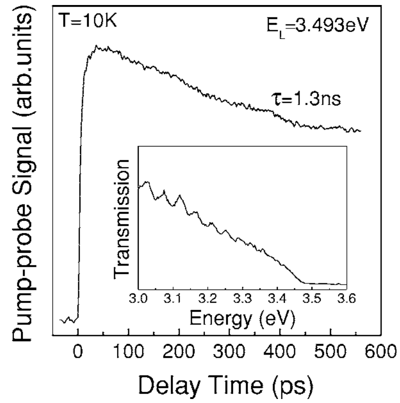

FIG. 2. Time-resolved pump-probe reflectivity taken from *p*-type GaN epilayer (#1) at 10 K. The laser energy is $E_L=3.493$ eV. Inset is the transmission spectrum at 10 K.

we can safely rule out the possibility that the observed spin precession in *p*-GaN come from electrons. It should come from holes in the valence bands A, B or C of GaN. Based on the following discussions, we identify that the observed spin precession come from holes in the valence band B.

According to group theory,[17] the transverse g factor for A holes is equal to zero, i.e., $g_{\perp}^{A}=0$, so we can rule out that the observed spin precession comes from holes in the valence band A.

Figure 4 shows the TRKR curves at $B=7$ T for different laser energies. The spectral linewidth of the laser we used is ∼38 meV. The energy difference between bands A and B is 6 meV, and 37 meV between bands B and C.[18] When the central energy of the laser pulses is tuned at 3.446 eV which is about 20 meV lower in energy than the measured GaN band gap ($E_g=3.47$ eV, inset of Fig. 2), the valence band C cannot be excited. However, the observed oscillations yield $g_{\parallel}=2.1\pm0.1$, which is, within the experimental error, the same as the value measured with laser energy at 3.482 eV. From this, we may conclude that the observed spin precession originates from holes in the valence band B. Campo

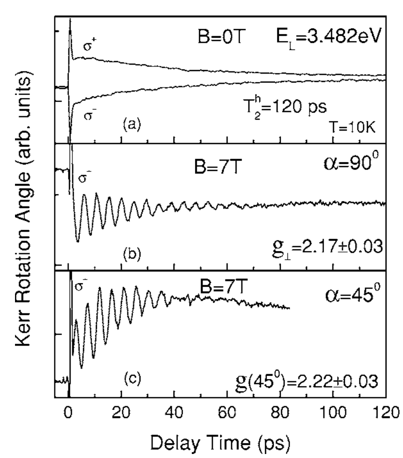

FIG. 3. TRKR taken from *p*-type GaN epilayer (#1) at 10 K and (a) $B=0$; (b) $B=7$ T at $\alpha=90°$; (c) $B=7$ T at $\alpha=45°$. The laser energy is $E_L=3.482$ eV and the pump beam is either σ^+ or σ^- polarized.

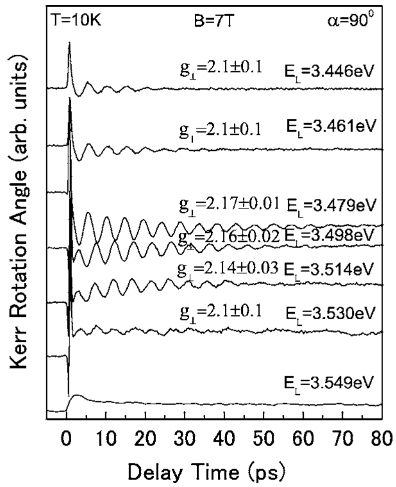

FIG. 4. TRKR taken from p-type GaN epilayer (#1) at 10 K and B=7 T for different central energies of laser pulses.

et al. measured the longitudinal g factor for the valence band B by means of magnetocircular dichroism, i.e., $g_{\parallel}^{h}=2.1\pm0.3$,[19] which is the same as our measured value within the experimental error. This further supports our conclusion that the observed spin precession comes from the holes in the valence band B.

When the laser energy is tuned from 3.446 eV to 3.530 eV, bands A, B, C are all excited and hole spins are created in all three bands. In principle, we should be able to observe the spin precession of holes from the valence band C. However, all TRKR curves show only one oscillation period and give the same $g_{\perp}$ value as that for the valence band B within the experimental errors. As the holes excited in the valence band C is 37 meV higher in energy than B holes, the C holes can rapidly relax into the valence band B, which may result in shorter spin coherence time of C holes. When the laser energy is tuned higher than 3.530 eV, the spin precession of B holes disappears. This can be due to the fast spin relaxation of holes enhanced with energy relaxation processes, or the poorer initial spin polarization under off-resonant excitation. When the laser energy is tuned lower than 3.446 eV, no spin precession is observed as the photoexcitation of holes does not occur any longer.

The TRKR curves in Fig. 3(a) shows only one decay constant except one sharp peak around zero time delay.[16] We can determine the spin coherence time of B holes $T_2^{h*}=120$ ps, which is 10 times shorter than the electron spin coherence time 2 ns in n-type GaN (see Fig. 2). Such a short hole spin coherence time is expected due to the decoherence induced by the valence-band mixing.[8] Similar to electron spins,[6] the hole spin coherence time is magnetic-field dependent, and it reduces to about 50 ps at magnetic field of 7 T (see Figs. 3 and 4).

Circularly polarized pump beam can excite both electron and hole spins. In principle, the spin precession of electrons and holes can be observed simultaneously from the same sample, and a beat pattern is expected. In this case, the Kerr rotation angle $\theta_K(t)$ can be expressed as

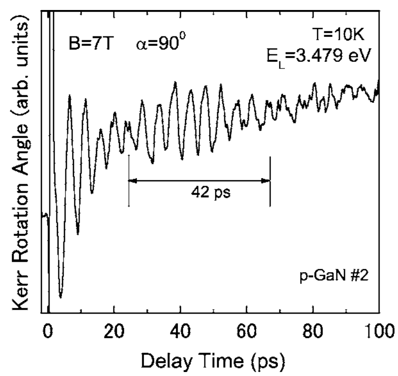

FIG. 5. TRKR taken from p-type GaN epilayer (#2) at 10 K and B=7 T. A beat pattern with a beat period of about 42 ps appears.

$$\theta_K(t) \propto g^e\langle S_0^e\rangle e^{-t/T_2^e}\cos(2\pi\nu_e t) + g^h\langle S_0^h\rangle e^{-t/T_2^h}\cos(2\pi\nu_h t), \quad (2)$$

where $\nu_e=g^e\mu_B B/h$ and $\nu_h=g^h\mu_B B/h$ are spin precession frequencies of electrons and holes, $\langle S_0^e\rangle$ and $\langle S_0^h\rangle$ are the total spins of photoexcited electrons and holes, T_2^e and T_2^h are spin coherence time of electrons and holes, and t is the delay time. Assuming $\langle S_0^e\rangle=\langle S_0^h\rangle$, $t \ll T_2^e, T_2^h$, and taking $g^e \simeq g^h$ with the same sign, Eq. (2) yields

$$\theta_K(t) \propto g^e\langle S_0^e\rangle\cos[\pi(\nu_e-\nu_h)t]\cos[\pi(\nu_e+\nu_h)t]. \quad (3)$$

The beat period is $T_{\mathrm{beat}}=1/|\nu_e-\nu_h|=h/[|g^e-g^h|\mu_B B]$, which decreases with increasing the magnetic fields. From the g factor values we measured, we can estimate the beat period to be 46 ps at B=7 T. We indeed observe such a beat pattern with beat period about 42 ps in one piece of p-GaN sample as shown in Fig. 5.[20] The envelope curve with a maximum at t=0 follows a cosine function and proves that g^h has the same sign as g^e [see Eqs. (2) and (3)].[21] However, we observe only one and a half beat periods, indicating that the spin coherence time of electrons as well as that of B holes are in the same range as the beat period (42 ps). Compared with 2 ns in n-GaN, such a short spin coherence time of electrons in p-GaN suggests that holes induces significant decoherence to electrons via electron-hole scatterings.

The g-factor anisotropy of B holes is $\Delta g^B=g_{\parallel}^B-g_{\perp}^B=0.10$, indicating nearly isotropic g factor for the valence band B, in contrast to the strongly anisotropic g factor of the valence band A. Since $g_{\perp}^A=0$ according to group theory, the spin precession of A holes cannot take place, and thus cannot be observed [see Figs. 3(b) and 4] when the magnetic field is perpendicular to the c-axis ($\alpha=90°$). However, we are not clear why the spin precession of A holes cannot be observed either when $\alpha=45°$, which is worth further investigation. We notice that the g-factor anisotropy of B holes is very close to that of holes bound to the Mg acceptor measured by EPR and optically detected magnetic resonance experiments.[10–13] This suggests that the electronic states of the valence band B play a major role in the wave function of the Mg acceptor in wurtzite GaN. The mixing of states can be caused by, e.g., the random strain fields in samples with high density of points defects and dislocations.[12]

In conclusion, we have studied the spin dynamics of electrons and holes in Si-doped (n-type) and Mg-doped (p-type) wurtzite GaN by the TRKR technique. In n-type GaN, we observe the spin precession of electrons in the conduction band and the spin coherence time of electrons is measured to be 2 ns. The g factor anisotropy of electrons is determined to be $g^e_\perp = 1.9492 \pm 0.0008$ and $g^e_\parallel = 1.9508 \pm 0.0008$. In p-type GaN, we observe the spin precession of holes from the valence band B and the spin coherence time of B holes is measured to be 120 ps. The g factor anisotropy of B holes is determined to be $g^h_\perp = 2.17 \pm 0.03$ and $g^h_\parallel = 2.27 \pm 0.03$. The beat induced by the spin precession of electrons and holes is also observed in p-type GaN.

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*Electronic address: cyhu03@yahoo.com
†Electronic address: oono@riec.tohoku.ac.jp
‡Electronic address: ohno@riec.tohoku.ac.jp

[1] T. Dietl, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, Science **287**, 1019 (2000).
[2] *Semiconductor Spintronics and Quantum Computation*, edited by D. D. Awschalom, N. Samarth, and D. Loss (Springer-Verlag, Berlin, 2002).
[3] J. J. Baumberg, D. D. Awschalom, N. Samarth, H. Luo, and J. K. Furdyna, Phys. Rev. Lett. **72**, 717 (1994).
[4] J. M. Kikkawa and D. D. Awschalom, Phys. Rev. Lett. **80**, 4313 (1998).
[5] J. M. Kikkawa, I. P. Smorchkova, N. Samarth, and D. D. Awschalom, Science **277**, 1284 (1997).
[6] 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).
[7] K. Kuroda, Y. Yabushita, T. Kosuge, A. Tackeuchi, K. Taniguchi, T. Chinone, and N. Horio, Appl. Phys. Lett. **85**, 3116 (2004).
[8] X. Marie, T. Amand, P. Le Jeune, M. Paillard, P. Renucci, L. E. Golub, V. D. Dymnikov, and E. L. Ivchenko, Phys. Rev. B **60**, 5811 (1999); X. Marie, T. Amand, J. Barrau, P. Renucci, P. Lejeune, and V. K. Kalevich, *ibid.* **61**, 11065 (2000).
[9] R. Winkler, S. J. Papadakis, E. P. De Poortere, and M. Shayegan, Phys. Rev. Lett. **85**, 4574 (2000).
[10] D. M. Hofmann, W. Burkhardt, F. Leiter, W. von Förster, H. Alves, A. Hofstaetter, B. K. Meyer, N. G. Romanov, H. Amano, and I. Akasaki, Physica B, **273–274**, 43 (1999).
[11] E. R. Glaser, W. E. Carlos, G. C. B. Braga, J. A. Freitas, Jr., W. J. Moore, B. V. Shanabrook, R. L. Henry, A. E. Wickenden, D. D. Koleske, H. Obloh, P. Kozodoy, S. P. DenBaars, and U. K. Mishra, Phys. Rev. B **65**, 085312 (2002).
[12] E. R. Glaser, J. A. Freitas, Jr., B. V. Shanabrook, D. D. Koleske, S. K. Lee, S. S. Park, and J. Y. Han, Phys. Rev. B **68**, 195201 (2003).
[13] R. Stepniewski, A. Wysmolek, M. Potemski, K. Pakua, J. M. Baranowski, I. Grzegory, S. Porowski, G. Martinez, and P. Wyder, Phys. Rev. Lett. **91**, 226404 (2003).
[14] The epilayers were provided by Nitride Semiconductors Co. Ltd. The carrier densities were evaluated by room-temperature Hall measurements.
[15] W. E. Carlos, J. A. Freitas, Jr., M. A. Khan, D. T. Olson, and J. N. Kuznia, Phys. Rev. B **48**, 17878 (1993).
[16] The sharp peaks around delay time $t=0$ (Figs. 1 and 3–5) are coherent artifacts which has nothing to do with spin.
[17] D. G. Thomas and J. J. Hopfield, Phys. Rev. **128**, 2135 (1962).
[18] G. D. Chen, M. Smith, J. Y. Lin, H. X. Jiang, Su-Huai Wei, M. Asif Khan, and C. J. Sun, Appl. Phys. Lett. **68**, 2784 (1996).
[19] J. Campo, M. Julier, D. Coquillat, J. P. Lascaray, D. Scalbert, and O. Briot, Phys. Rev. B **56**, R7108 (1997).
[20] The spin coherence time of holes for the sample piece #2 is a little longer than that for the sample piece #1 due to the inhomogeneity of the wafer. This gives a chance to observe the beat pattern between spin precession of electrons and holes in the sample piece #2.
[21] As the spin-orbit interaction is much smaller than the band gap energy in GaN, the g factor of conduction band electrons is close to 2 and has positive sign. So the g factor of B holes has a positive sign (or the g factor of electrons in the valence band B has negative sign, in accordance with Ref. 19).