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Exciton spin relaxation in GaN observed by spin grating experiment

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The authors studied the exciton spin relaxation of bulk GaN by creating spin polarization gratings using degenerate four-wave mixing spectroscopy. The spectrally resolved analysis achieved with this technique facilitated the direct evaluation of spin polarizations in the individual excitons (*A* and *B* excitons). The spin polarizations for each exciton decay very quickly ($\tau_s \sim 1$ ps) at low temperatures. Moreover the τ_s is faster than the dephasing time T_2 throughout the measured temperature range, suggesting the existence of fast intrinsic spin relaxation processes, which can be attributed to a large exchange constant characterized in GaN. © 2007 American Institute of Physics. [DOI: 10.1063/1.2430402]

Because of the technological importance of spin-based electronics, so-called spintronics,¹ considerable effort has recently been devoted to understanding the optically induced spin polarization in GaN and related alloys, both experimentally²⁻⁵ and theoretically.⁶⁻⁹ A noteworthy issue is the significant variation in the spin relaxation time obtained in previous experiments; in Ref. 2, a long electron spin coherence of up to 20 ns was observed in Si-doped GaN by using time-resolved Faraday rotation measurements, while short (subpicosecond) excitonic-spin relaxation times were observed in a GaN epilayer by using spin-dependent transient reflectivity change measurements.³ Although theoretical work has suggested a significant influence of the sample quality on the spin dynamics in semiconductors,⁷ the mechanism that causes the difference between these two results is still unclear.

In this work, we studied exciton spin relaxation in bulk GaN by using spectrally resolved four-wave mixing (SR-FWM) measurements. Strikingly fast spin relaxations, which were even faster than the dephasing times T_2 , were observed for each exciton resonance at low temperatures. The relaxation time in an *A* exciton (X_A) decreases with increasing temperature, and then reaches the experimental resolution limit above 100 K. The results can be understood in terms of an excitonic effect in GaN.

Figure 1(a) is a schematic illustration of the experiment. In order to evaluate the spin lifetime in the spectral domain, we employed a three-pulse FWM measurement.¹⁰⁻¹² A frequency doubled, mode-locked Ti:sapphire laser which produced a pulse width of ~ 200 fs (spectral width of ~ 13 meV) at a repetition rate of 76 MHz was employed as an excitation light source. Two simultaneous excitation pulses and one probe pulse with a controlled time delay were focused at the same point on the sample surface using a lens ($f=200$ mm). These three pulses have almost the same power of about 0.3 mW. The pump pair consisting of cross-linear (colinear) polarizations of wave vectors \mathbf{k}_1 and \mathbf{k}_2 forms an exciton spin polarization (population) grating [Fig. 1(b)] which mainly decays via the exciton spin relaxation (recombination) process.¹³ A third probe pulse of wave vector \mathbf{k}_3 diffracts off this grating to produce the FWM sig-

nal in the $\mathbf{k}_4 = \mathbf{k}_3 \mp \mathbf{k}_2 \pm \mathbf{k}_1$ direction, which was selected spatially by an iris and resolved spectrally by a 30-cm-long monochromator with a charge coupled device detector. The spectral resolution is 0.7 meV.

Here we briefly describe the excitonic-spin polarization in wurtzite GaN. Figure 1(c) shows an overview of the energy structure of an electron and holes for the *A* and *B* excitons (X_A and X_B) in GaN based on the general group theory. The direct products of the lowest conduction band with Γ_7 symmetry and the topmost of the valence band with Γ_9 give X_A with $\Gamma_5(x,y)$ and Γ_6 symmetry, where only $\Gamma_5(x,y)$ is optically allowed. On the other hand, X_B arises from the second valence band with Γ_7 symmetry and consists of two optically allowed states $\Gamma_1(z)$ and $\Gamma_5(x,y)$. Both in X_A and X_B , $\Gamma_5(x,y)$ is decomposed into two opposite circularly polarized spin states: $|+1\rangle$ and $|-1\rangle$.

In our study, we investigated a 70- μm -thick freestanding GaN sample, which was grown by the two-flow metal-organic chemical vapor deposition method using the lateral

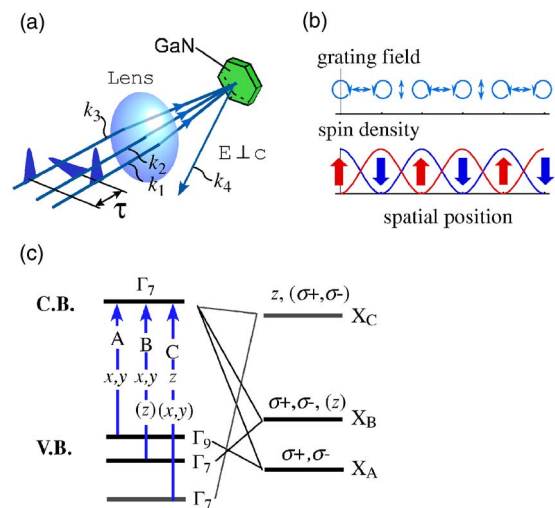


FIG. 1. (Color online) (a) Excitation scheme for generating a spin polarization grating induced by cross-linearly polarized pump pulses with wave vectors \mathbf{k}_1 and \mathbf{k}_2 . The pump pulses were mechanically delayed with respect to the third probe pulse with wave vector \mathbf{k}_3 . (b) Spatial distribution of polarization in the cross-polarized excitation (upper) and corresponding spin density distribution (lower). (c) Schematic of energy bands at the Γ point and the internal structure of excitons in wurtzite crystals based on the general group theory.

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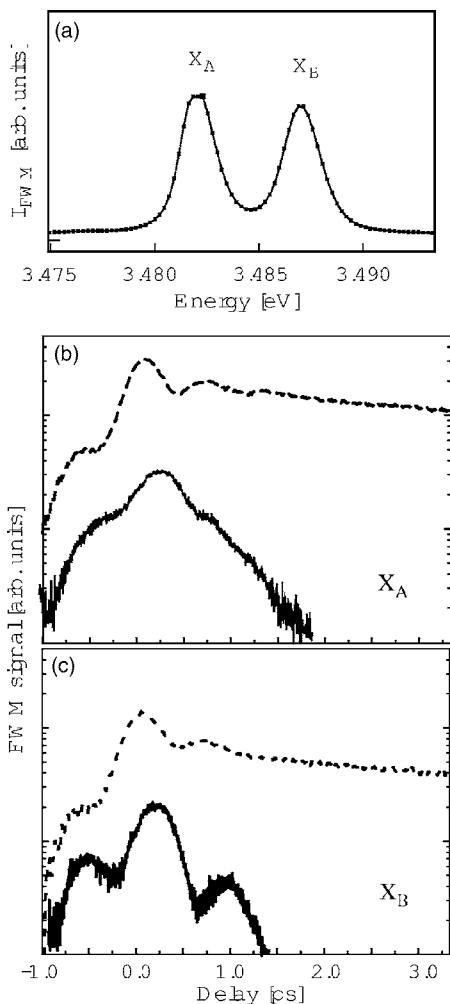


FIG. 2. (a) Typical FWM spectrum of the sample at delay time $\tau=0$ s. The solid line is a Lorentzian fitting. SRFWM for (b) X_A and (c) X_B . The solid and dashed lines represent the spin polarization transient and the population transient, respectively. The results were obtained at 10 K.

epitaxial overgrowth technique. We should point out that the residual carrier concentration was of the order of 10^{14} cm^{-3} , indicating a small quantity of residual impurities. The sample characteristics are described in detail in Ref. 14.

A typical SRFWM spectrum at zero time delay is shown in Fig. 2(a), which was obtained for the pump pulses in a cross-linearly polarized configuration. In the spectrum, the lower- and higher-energy peaks can be identified as X_A and X_B , respectively. The exciton energies obtained from the Lorentzian fitting to the data were 3.4809 and 3.4862 eV, respectively. There was an energy separation of ~ 5 meV between the two excitons. This separation is smaller than that of the excitation laser spectrum and so means that it is impossible to achieve the selective excitation of individual exciton levels that is required if we are to obtain spin-dependent transient measurements.^{3,4} In the two-pulse FWM measurements, the T_2 values of each exciton, where we assume the homogeneous broadening, are estimated to be 1.6 and 1.7 ps, respectively. These values are consistent with the linewidths in the spectrum.¹⁵ This consistency and the relatively long T_2 account for the high crystal quality of our thick sample.

Figures 2(b) and 2(c) display the results of three-pulse SRFWM for X_A and X_B , respectively, at the lowest temperature ($T=10$ K). An intense signal with a long decay was

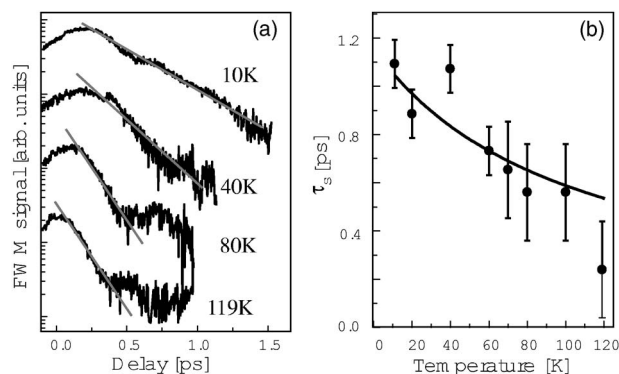


FIG. 3. (a) Spin polarization transients of X_A at various temperatures. (b) Plots of decay time obtained from a least-square fit to the data. The solid curve is the result of fitting using a model described in the text.

observed by using colinearly polarized pump excitation, in which the fast decay component corresponds to the dephasing process, while the subsequent slow decay corresponds to a lifetime of the order of several tens of picoseconds. In contrast, the FWM signal observed in the cross-linear configuration shows a very rapid decay. With this geometry, the signal reflects the decay of the periodic pattern of the polarization grating [Fig. 1(b)]. A small oscillatory component superimposed on the exponential decay originates from the remnant of the two-pulse FWM signals (e.g., $2\mathbf{k}_2 - \mathbf{k}_1$) after spatial filtering, and we disregard this contribution when evaluating the decay. By fitting $\exp(-2\tau/\tau_s)$ to the data, we obtained a typical spin relaxation time of $\tau_s=1.2$ ps at X_A resonance at 10 K. On the other hand, the oscillation is dominant at X_B , indicating a small amount of spin polarization. The reduced polarization compared with X_A is due to the excited state nature of X_B , which provides an additional relaxation channel toward the lowest X_A state.

Figure 3 (a) shows semilog plots of the SRFWM intensities of X_A at various temperatures. The pump polarizations were set so that they crossed. In Fig. 3(b), we plot τ_s obtained by a single exponential fit to the data. The τ_s value decreases monotonically with increasing temperature, and finally becomes comparable to the pulse duration above $T \sim 100$ K. This temperature dependence of τ_s clearly indicates the scattering process contribution, which can be evaluated by comparison with the temperature dependence of T_2 . For this purpose, we employ the fitting function expressed as $\tau_s(T) = 2\hbar / (\alpha_0 + \beta_{\text{ph}}T)$, where α_0 and β_{ph} correspond to the homogeneous broadening Γ_h at zero temperature for $T_2 = 2\hbar / \Gamma_h$ and to the exciton-acoustic phonon interaction coefficient, respectively. From a least-square fitting procedure, we have obtained the values $\alpha_0 = 1.1$ meV and $\beta_{\text{ph}} = 11 \mu\text{eV/K}$. The β_{ph} value is comparable to that for T_2 in GaN reported by several authors,¹⁶ while α_0 is very large, meaning that τ_s is faster than T_2 at zero temperature. These results suggest that there must be intrinsic fast relaxation processes additional to the exciton-phonon-scattering-induced relaxation. This will be discussed below.

Similar fast spin relaxation with a τ_s of 0.47–0.25 ps at 150–225 K has been observed in a GaN epilayer by using spin-dependent transient reflectivity change measurements.³ In contrast, in Ref. 2, the electron spin coherence in n -doped GaN becomes as long as ~ 20 ns at 5 K. This noticeable difference might result from the relatively low quality of the former sample since defects and dislocations originating

from the substrate will not relax in such a thin sample. Although GaN essentially has a high defect density, the observed subpicosecond spin relaxation time in our thick sample supports the generality of a fast τ_s in terms of excitonic transitions, which in turn indicates that the difference in spin relaxation time can be attributed to the difference between electron spin and exciton spin relaxations.

Usually, electron/hole spin is affected by a spin-orbit (SO) interaction:¹⁷ a strong SO coupling results in a large electron spin splitting at large \mathbf{k} , where scattering caused by impurities and phonons gives rise to efficient spin relaxation. Since GaN exhibits a small SO splitting $\Delta_{\text{SO}} \approx 13$ meV, which is smaller than GaAs ($\Delta_{\text{SO}} \approx 350$ meV), a long-lived electron spin is predicted⁸ and accounts well for the electron spin experiments on doped GaN.² On the other hand, in an electron-hole picture, excitonic exchange interactions provide an additional spin relaxation channel that accounts for the simultaneous flip between electrons and holes, known as the Bir-Aronov-Pikus mechanism.¹⁸ Optical experiments reveal a large spin exchange constant $\gamma = 0.6$ meV of GaN,¹⁹⁻²¹ which is about three times that of GaAs ($\gamma = 0.25$ meV). Since τ_s is inversely proportional to the square of the exchange energy, we can expect an order of magnitude reduction in τ_s .²² Moreover, strikingly fast hole spin relaxation was theoretically predicted in GaN due to the weak SO coupling and the nearly degenerate large effective mass.⁸ In this case, the exciton spin relaxation time shortens as a result of the large exchange interaction.

It is important to note that τ_s is even faster than T_2 evaluated by two-pulse FWM for temperatures up to around 120 K. Moreover, the β_{ph} value for τ_s is nearly identical to that for T_2 , indicating that the contribution of motional narrowing is small. The fact that τ_s is faster than T_2 indicates that scattering-induced relaxations are not dominant for τ_s , although they provide a temperature dependent contribution. As a consequence, we conclude that the fast τ_s is determined by the intrinsic spin-flip processes of GaN excitons, which are possibly caused by the large exchange interaction.

In summary, we have investigated the three-pulse FWM response of exciton spin polarization in a high-quality bulk GaN. The spin relaxation times in each of the exciton states were estimated to be ~ 1 ps at 10 K. Based on a disagreement from long-lived electron spin, we argue that the short spin relaxation times observed at exciton resonance originate from the excitonic interaction. A large exchange interaction

in GaN provides a plausible explanation for this efficient spin relaxation.

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- ¹*Semiconductor Spintronics and Quantum Computation*, edited by D. D. Awschalom, N. Samarth, and D. Loss (Springer, Berlin, 2002).
- ²B. 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 (2001).
- ³T. Kuroda, T. Yabushita, T. Kosuge, A. Tackeuchi, K. Taniguchi, T. Chinone, and N. Horio, *Appl. Phys. Lett.* **85**, 3116 (2004).
- ⁴S. Nagahara, M. Arita, and Y. Arakawa, *Appl. Phys. Lett.* **86**, 242103 (2005).
- ⁵C. Y. Hu, K. Morita, H. Sanada, S. Matsuzaka, Y. Ohno, and H. Ohno, *Phys. Rev. B* **72**, 121203(R) (2005).
- ⁶D. Jena, *Phys. Rev. B* **70**, 245203 (2004).
- ⁷S. Krishnamurthy, M. V. Schilfgaarde, and N. Newman, *Appl. Phys. Lett.* **83**, 1761 (2003).
- ⁸Z. G. Yu, S. Krishnamurthy, M. V. Schilfgaarde, and N. Newman, *Phys. Rev. B* **71**, 245312 (2005).
- ⁹P. H. Song and K. W. Kim, *Phys. Rev. B* **66**, 035207 (2002).
- ¹⁰A. R. Cameron, P. Riblet, and A. Miller, *Phys. Rev. Lett.* **76**, 4793 (1996).
- ¹¹O. Ikeuchi, S. Adachi, H. Sasakura, and S. Muto, *J. Appl. Phys.* **93**, 9634 (2003).
- ¹²S. Adachi, Y. Takagi, J. Takeda, and K. A. Nelson, *Opt. Commun.* **174**, 291 (2000).
- ¹³Since the diffracted signals are proportional to the depth of the generated grating, exciton (spin) diffusion contributes to the signal decay as well as the recombination (spin relaxation) process. In these experiments, the grating pitch is so large that the diffusion effect cannot affect the grating decay on the time scale of interest.
- ¹⁴K. Torii, T. Deguchi, T. Sota, K. Suzuki, S. Chichibu, and S. Nakamura, *Phys. Rev. B* **60**, 4723 (1999); T. Deguchi, D. Ichiryu, K. Toshikawa, K. Sekiguchi, T. Sota, R. Matsu, T. Azuhata, M. Yamaguchi, T. Yagi, S. Chichibu, and S. Nakamura, *J. Appl. Phys.* **86**, 1860 (1999).
- ¹⁵Since the resonance linewidth of ~ 2 meV is only 2.5 times that for a homogeneous case (linewidth of 0.8 meV from $T_2 \sim 1.6$ ps), the system is close to exhibiting homogeneous broadening.
- ¹⁶K. Hazu, A. Shikanai, T. Sota, K. Suzuki, S. Adachi, S. F. Chichibu, and T. Mukai, *Phys. Rev. B* **65**, 195202 (2002); S. Pau, J. Kuhl, F. Scholz, V. Haerle, M. A. Khan, and C. J. Sun, *Appl. Phys. Lett.* **72**, 557 (1998); A. J. Fischer, W. Shan, G. H. Park, J. J. Song, D. S. Kim, D. S. Yee, R. Horning, and B. Goldenberg, *Phys. Rev. B* **56**, 1077 (1997).
- ¹⁷R. J. Elliott, *Phys. Rev.* **96**, 266 (1954).
- ¹⁸G. L. Bir, A. G. Aronov, and G. E. Pikus, *Zh. Eksp. Teor. Fiz.* **69**, 1382 (1975) [*Sov. Phys. JETP* **42**, 705 (1976)].
- ¹⁹M. Julier, J. Campo, B. Gil, J. P. Lascaray, and S. Nakamura, *Phys. Rev. B* **57**, R6791 (1998).
- ²⁰P. P. Paskov, T. Paskova, P. O. Holtz, and B. Monemar, *Phys. Status Solidi A* **190**, 75 (2002).
- ²¹Y. Toda, S. Adachi, Y. Abe, K. Hoshino, and Y. Arakawa, *Phys. Rev. B* **71**, 195315 (2005).
- ²²M. Z. Maialle, *Phys. Rev. B* **54**, 1967 (1996).