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Spin splitting in modulation-doped $Al_xGa_{1-x}N/GaN$ heterostructures

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We have studied the electronic properties of $Al_xGa_{1-x}N/GaN$ heterostructures by using Shubnikov–de Haas (SdH) measurement. Two SdH oscillations were detected on the samples of $x=0.35$ and 0.31, due to the population of the first two subbands with the energy separations of 128 and 109 meV, respectively. For the sample of $x=0.25$, two SdH oscillations beat each other, probably due to a finite zero-field spin splitting. The spin-splitting energy is equal to 9.0 meV. The samples also showed a persistent photoconductivity effect after illuminating by blue light-emitting diode.

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The spin splitting of conduction band in the inversion layer of semiconductor heterostructures has been investigated both theoretically[1,2] and experimentally[3,4] for decades. The intrinsic splitting is caused by the confinement potential of a two-dimensional electron gas (2DEG) in the present of an interface electric field. Two competing mechanisms were used to interpret the zero-field spin splitting; one is the inversion-asymmetry-induced bulk k^3 term and the other is a linear interface spin-orbit interaction.[1] The calculated spin splitting in GaAs/AlAs due to the inversion-asymmetry-induced k^3 term is about 4 meV,[2] while the theoretical spin splitting arisen from the linear spin-orbit interaction is smaller than 1 meV.[5] However, the experimental spin splitting determined from the Shubnikov–de Haas (SdH) measurement on InGaAs/InAlAs is about 1.5~2.5 meV.[4] The value is still unclear to identify the mechanism of the spin splitting. In order to distinguish these two mechanisms, we need a sample with a large interface electric field to enhance the inversion asymmetry, and its spin splitting exceeds 4 meV, which is ascribed to the inversion-asymmetry-induced k^3 term. Because the $Al_xGa_{1-x}N/GaN$ heterostructures have a large conduction band discontinuity and a strong piezoelectric effect, a high internal electric field is generated at the interface. Therefore the $Al_xGa_{1-x}N/GaN$ heterostructure is a suitable material to evaluate the spin-splitting effect. Shubnikov–de Haas effect on the $Al_xGa_{1-x}N/GaN$ heterostructures has been studied for varied Al compositions from $x=0.1$ to 0.18 by Wang *et al.*,[6] and for different barrier thickness with $x=0.22$ by Jiang *et al.*[7] There were two SdH oscillations observed in both papers. Jaing *et al.* attributed the additional oscillation to the second subband population, while Wang *et al.* did not discuss about it. However, the secondary SdH oscillation in Wang's paper showed apparently different characteristics from that in Jiang's paper. The conduction-band bending depends strongly on charge transferring and polarization field for the piezoelectric materials such as AlGaN/GaN or InAlAs/InGaAs. Since the greater Al composition (x) yields a larger conduction-band offset and hence more electrons are transferred into the GaN triangular potential well. A high carrier concentration is achieved at the interface and the second subband is consequently occupied. In addition to the charge transferring, a strong macroscopic polarization field is generated at the $Al_xGa_{1-x}N/GaN$ interface due to the piezoelectric effect. A linear increase of the polarization field with the Al composition was reported.[8] The theoretical calculation showed that the carrier concentration due to the piezoelectric field depends on the Al composition of $Al_xGa_{1-x}N$ layer as well.[9] If the high electric field generates a large inversion-asymmetry-induced spin splitting in the $Al_xGa_{1-x}N/GaN$ heterostructure, then we should be able to detect the splitting on the SdH measurement.

Three samples of the modulation-doped $Al_xGa_{1-x}N/GaN$ heterostructure were used in this study. The samples were grown on a sapphire (Al_2O_3) substrate by metal-organic chemical vapor deposition and consisted of 1~2 μm GaN 100 Å undoped-$Al_xGa_{1-x}N$ spacer and 200 Å Si-doped n-$Al_xGa_{1-x}N$ barrier [see the inset in Fig. 1(b)]. Al composition changes from $x=0.35$ to 0.25 and Si-doping concentration varies from 5×10^{18} to $1.7\times10^{17}\ \mathrm{cm}^{-3}$. The Hall carrier concentration (n_H) and mobility (μ_H) of the samples were determined by van der Pauw Hall measurement and are shown in Table I. The SdH measurement was performed on Hall bar samples, and the data were taken with equal spacing of $1/B$ (for the Fourier transformation purpose) from 0.5 to 12 T at the temperature of 1.2 K. The magnetoresistance R_{XX} oscillates with $1/B$ and the SdH frequency f_i depends only on the carrier concentration n_i for the ith subband: $f_i=hn_i/2e$. The energy difference between Fermi level E_F and the ith subband minimum E_i can be calculated by $E_F-E_i=\pi\hbar^2n_i/m^*$, where m^* is the effective mass and h is the

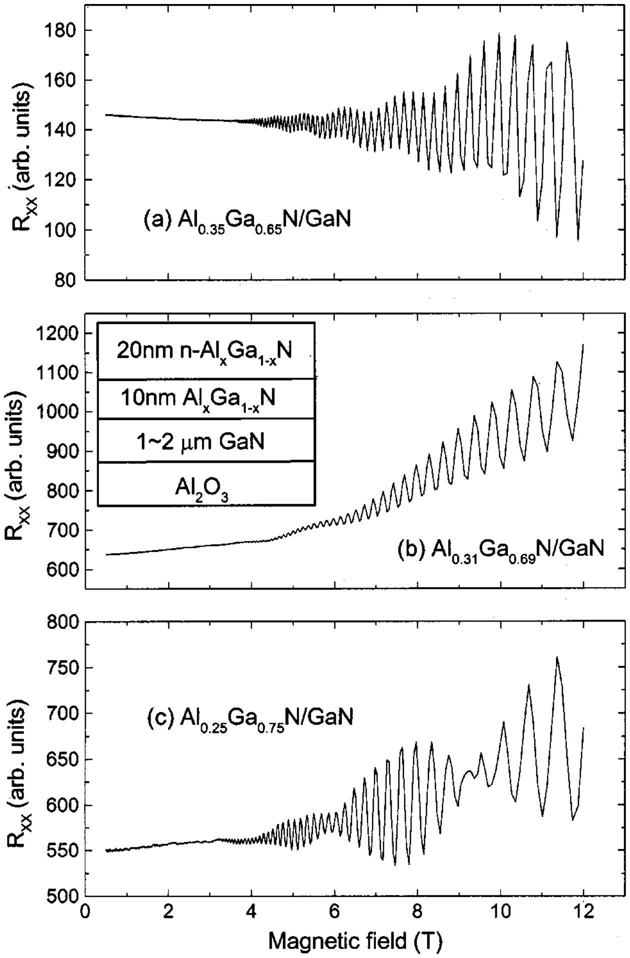

FIG. 1. The SdH oscillations after illuminating the samples at 1.2 K for about 780 s.

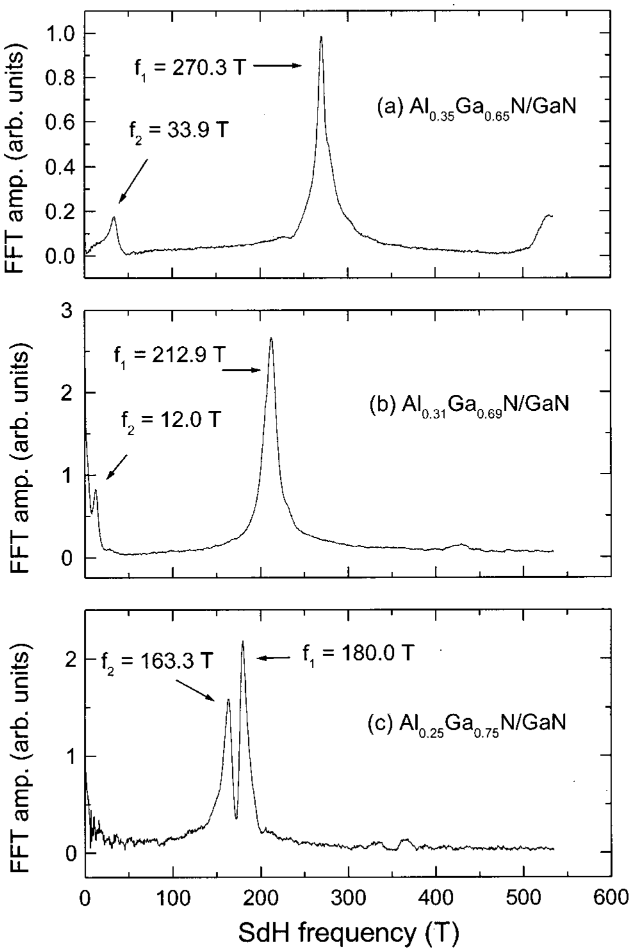

FIG. 2. The FFT spectra of the SdH oscillations in Fig. 1.

Planck's constant.[10] The SdH oscillations were observed in all of the samples. The illumination of the samples by a blue light emitting diode at low temperature increases the carrier concentration and the increase is persistent after the removal of the light (i.e., a persistent photoconductivity effect). Figure 1 shows the plots of R_{XX} versus the magnetic field after illuminating the samples for about 780 s. The fast Fourier transformations (FFT) of these SdH data are shown in Fig. 2. The SdH frequencies of the primary (f_1) and the secondary (f_2) oscillations are determined from the FFT spectrum. In Figs. 2(a) and 2(b), if the two SdH frequencies are due to the lowest two subbands, the carrier concentrations of the two subbands obtained from the two SdH frequencies are 1.31×10^{13} and 1.6×10^{12} cm^{-2} for sample 1, and 1.03×10^{13} and 5.8×10^{11} cm^{-2} for sample 2, respectively. It is reasonable, because sample 1 has a higher carrier concentration due to the larger x and higher Si-doping level. If we assume that the effective mass for the two subbands is about the same, $m^*=0.215m_0$, the energy separation between the two subbands is $E_2-E_1=128$ meV for sample 1, and 109 meV for sample 2. Garrido *et al.* calculated the energy separation of the first and second subbands in Al$_{0.25}$Ga$_{0.75}$N/GaN heterostructure to be 112 meV.[8] The theoretical value is in good agreement with our experimental results. The small deviation probably arises from the different sample parameters (e.g., in Garrido's sample, $x=0.25$, the barrier thickness is 350 Å, the spacer is 30 Å, and the Si-doping level is 2×10^{18} cm^{-3}), which can affect the piezoelectric field and charge transferring. Therefore, in samples 1 and 2, electrons transfer from the barrier into the 2DEG and start to populate the second subband due to the high carrier concentration. In sample 3,

TABLE I. The parameters of samples and their Hall carrier concentration and mobility determined by van der Pauw measurements at 10 K.

Sample	Al composition (x)	Si-doping concentration (cm^{-3})	n_H (cm^{-2})	μ_H (cm^2/V s)
1	0.35	5×10^{18}	1.51×10^{13}	2720
2	0.31	3×10^{18}	1.24×10^{13}	4009
3	0.25	1.7×10^{17}	8.96×10^{12}	6120

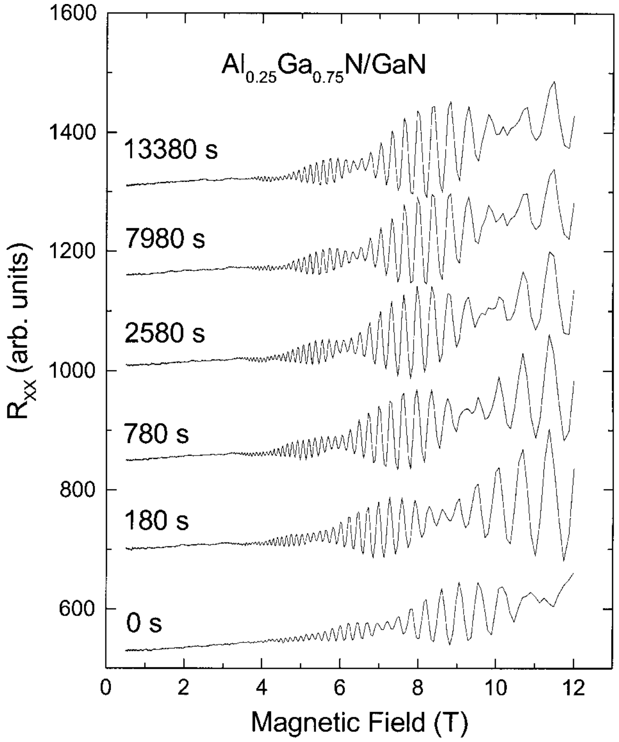

FIG. 3. The SdH oscillations of sample 3 for different illumination times at 1.2 K.

there are two SdH oscillations beating each other with two frequencies, $f_1=180.0$ and $f_2=163.3$ T [see Figs. 1(c) and 2(c)]. The beating effect still exists even after illuminating the sample for longer times. Figure 3 shows the plot of R_{XX} versus the magnetic field for different illumination times. The two frequencies, obtained from the FFT spectra of the SdH data in Fig. 3, increase with the illumination time (Fig. 4). The beat frequency ($f_{\mathrm{beat}}=f_1-f_2$) did not change obviously, but the nodes shift to a higher field. The beating effect implies that the two SdH oscillations have similar frequencies and comparable amplitudes. There are three possibilities to produce the beating: (i) the spin splitting of the first subband, (ii) two-subband occupation, and (iii) the second-channel located at the $Al_{0.25}Ga_{0.75}N$ barrier. The theoretical band calculation shows that most of the free electrons transfer from the n-$Al_{0.25}Ga_{0.75}N$ barrier to the triangular potential well due to the large conduction band offset.[8] Even if there reside some electrons in the barrier layer, the impurity scattering will reduce the mobility of the electrons and hence their contribution can be ignored. Therefore, the third possibility is unlikely to produce the beating effect on sample 3. If we assume that two subbands are occupied, the carrier concentrations obtained from SdH frequencies are $n_1=8.73\times10^{12}$ and $n_2=7.92\times10^{12}$ cm^{-2}, which yield the energy separation of 9.0 meV. The theoretical energy separation of the two lowest subbands is about 112 meV. The carrier concentrations are inconsistent with the Hall carrier concentration shown in Table I, and the energy separation disagrees with the calculated value of 112 meV. Therefore, the second possibility is probably not the case of sample 3. However, if the beating effect has arisen from the spin splitting of the first subband, the spin degeneracy factor of 2 needs to be removed and the relation of SdH frequency to the carrier concentrations of spin-up and spin-down electrons become, $f_\uparrow=hn_\uparrow/e$ and $f_\downarrow=hn_\downarrow/e$. Thus the carrier concentrations obtained from the two frequencies are $n_\uparrow=4.36\times10^{12}$ and $n_\downarrow=3.96\times10^{12}$ cm^{-2}. These values agree with the Hall carrier concentration in Table I. The energy difference (i.e., the zero-field spin splitting) becomes $E_\downarrow-E_\uparrow=2\pi\hbar^2(n_\uparrow-n_\downarrow)/m^*$. We therefore calculated the spin-splitting energy from the SdH frequencies to be $E_\downarrow-E_\uparrow=9.0$ meV.

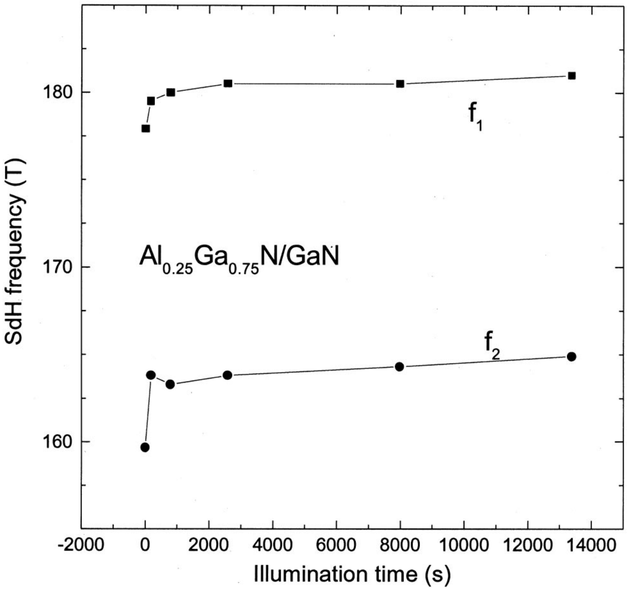

FIG. 4. The frequencies of SdH oscillations for sample 3 ($x=0.25$) versus illumination time.

What is the origin to generate the beating effect in sample 3? Das *et al.* observed the SdH beating effect in $In_xGa_{1-x}As/In_{0.52}Al_{0.48}As$ heterostructures and deduced a spin splitting of $1.5\sim2.5$ meV as the applied magnetic field approached zero.[4] The authors attributed the beating patterns to the finite zero-field spin splitting related to the inversion-asymmetry-induced bulk k^3 term.[1,2,4] Similar to the weak ferromagnetic electron system (e.g., $ZrZn_2$), the imbalance of the number of spin-up ($n_\uparrow$) and spin-down ($n_\downarrow$) electrons induces the spin-split Fermi surfaces at zero magnetic field and hence produces the beating oscillations on the Fermi surface measurements such as de Haas–van Alphen[11] or Shubnikov–de Haas effects.[4] Based on Das' model,[4] the amplitude of the beat pattern induced by spin-split Landau levels can be expressed as $A_{\mathrm{SdH}}\sim\cos\pi\nu$, where $\nu=\delta/\hbar\omega$, and δ is the total spin splitting of the Landau levels. If the total spin splitting δ is B dependent, it can be expanded on the cyclotron energy: $\delta=\delta_0+\delta_1\hbar\omega+\delta_2(\hbar\omega)^2+\cdots$, where δ_0 is the zero-field spin splitting and $\delta_1\hbar\omega$ is the linear-field splitting, and so on. Because $\delta_0=E_\downarrow-E_\uparrow=2\pi\hbar^2(n_\uparrow-n_\downarrow)/m^*$, a finite zero-field splitting δ_0 gives rise to a beat pattern against $1/B$ and the amplitude of the beat becomes $A_{\mathrm{SdH}}\sim\cos(\pi f_{\mathrm{beat}}/B)$ with a beat frequency, $f_{\mathrm{beat}}=f_\uparrow-f_\downarrow$, where we used $f_\uparrow=hn_\uparrow/e$ and $f_\downarrow=hn_\downarrow/e$. The linear-field term ($\delta_1\hbar\omega$) only contributes a constant phase to the beat pattern, $\pi\delta_1$. The higher-order terms, e.g., $\delta_2(\hbar\omega)^2$, will yield a beating effect with a B-dependent frequency, and occur in ferromagnetic materials whose electron exchange interaction is so strong that the spin-splitting Fermi surfaces depend greatly on the applied magnetic field (e.g., $f_{\uparrow}-f_{\downarrow}=184$ T in $ZrZn_2$).[11] However, in the semiconductor heterostructures, the spin splitting induced by the inversion asymmetry is small and the higher-order terms can be ignored. The amplitude becomes $A_{\mathrm{SdH}} \sim \cos \pi\nu = \cos(\pi f_{\mathrm{beat}}/B + \pi\delta_1)$. For example, in Das' paper the beating pattern was observed in sample *A*, and the last node of the beat pattern appears at $B = 0.873$ T (Fig. 2 in Ref. 4). The authors estimated the zero-field spin splitting by counting the half-integer values of $\nu(=1/2, 3/2, 5/2, \ldots)$ against $\hbar\omega$, and obtained $\delta_0 = 2.37$ meV (Fig. 4 in Ref. 4). However, since the amplitude of the beat A_{SdH} is against $1/B$, not $\hbar\omega$, the value of δ_0 extrapolated against $\hbar\omega$ in Ref. 4 is incorrect. We recalculated the beat frequency for the $In_xGa_{1-x}As/In_{0.52}Al_{0.48}As$ heterostructure from the plot of integer versus $1/B$ at the fields of the nodes in Fig. 2 of Ref. 4. The new plot is shown in the inset of Fig. 5, and the beat frequency is just equal to the slope $f_{\mathrm{beat}} = 0.925$ T. The linear fit in the new plot is much better than that in Fig. 4 of Ref. 4. Therefore, we recalculated the zero-field spin splitting for $In_xGa_{1-x}As/In_{0.52}Al_{0.48}As$ heterostructure, $\delta_0 = 1.16$ meV. In our case, the $Al_{0.25}Ga_{0.75}N/GaN$ heterostructure has a much higher piezoelectric effect than InGaAs/InAlAs.[12] The high piezoelectric field results in a greater band bending at the interface, and hence yields a larger spin-splitting energy $\delta_0 = 9.0$ meV. This can be checked by the tilt of the sample orientation with respect to the applied magnetic field. Since the Zeeman splitting energy is a function of total magnetic field B_{tot} while the SdH oscillation of 2DEG is a function of the perpendicular component of the applied field, i.e., $B \cos\theta$, where θ is the angle between the sample orientation and the applied field. The location of the nodes will move to higher fields whenever the Zeeman energy $(g^*\mu_B B_{\mathrm{tot}})$ becomes comparable to δ_0. We performed the SdH measurements on sample 3 with a rotating sample holder for θ from $-2°$ to $88°$. The results were shown against $B\cos\theta$ in Fig. 5, where the data of $\theta = 88°$ did not show in the plot. It is found that the locations of the nodes and SdH peaks are held nearly constant with $B\cos\theta$ for $\theta < 43°$. It is because in the field range (0.5–12 T) the Zeeman energy of sample 3 is still much smaller than δ_0. This occurs in the $In_xGa_{1-x}As/In_{0.52}Al_{0.48}As$ heterostructure for the field range from 0.15 to 1.0 T and $\theta < 70°$ (see Fig. 5 of Ref. 4). Therefore, we conclude that the beating effect on the SdH oscillations observed in sample 3 is probably caused by the finite zero-field spin splitting due to the inversion-asymmetry-induced bulk k^3 term.

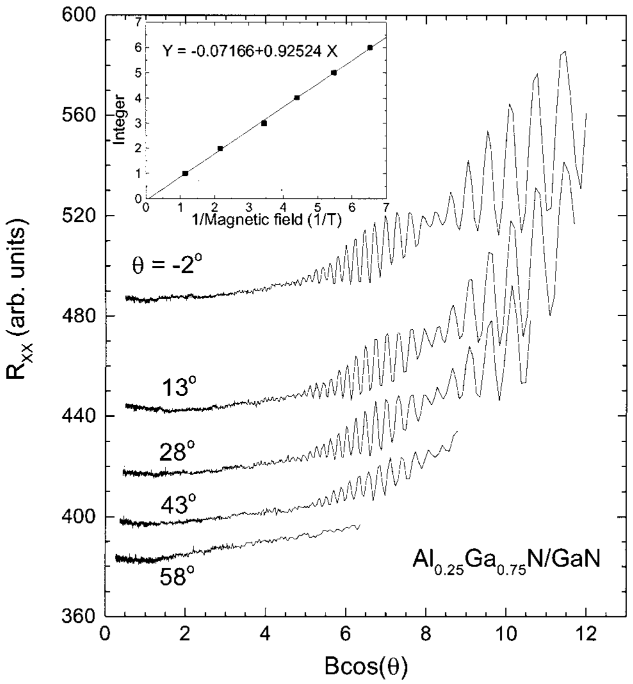

FIG. 5. The SdH oscillations of sample 3 for the angles: $\theta = -2°$, 13°, 28°, 43°, and 58°. The inset shows the plot of integer versus the inverse field of the nodes in Fig. 2 of Ref. 4. The beat frequency is obtained from the slope of the plot, $f_{\mathrm{beat}} = 0.925$ T.

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