Thermopower investigation of *n*- and *p*-type GaN

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A comparative investigation of the Hall effect, conductivity, and thermopower properties of molecular-beam-epitaxy-grown GaN is presented. In unintentionally doped *n*-type GaN, a negligible thermal activation of the thermopower is observed above 300 K. In as-grown GaN:Mg, a thermopower activation energy of 280 meV is observed at high temperatures, as well as a scattering factor $A=3$. At temperatures below 120 K, the Seebeck coefficient of *p*-type GaN changes sign and indicates *n*-type conductivity. These results show that hopping in the acceptor band contributes significantly to the electronic transport properties. After hydrogenation of GaN:Mg, both conductivity and thermopower have an activation energy of 520 meV, which is at variance with the presence of potential fluctuations in the material. This demonstrates that hydrogen passivates Mg-doped GaN by the formation of electrically inactive Mg-H complexes, in contrast to the formation of compensating H-related donors, which should lead to noticeable potential fluctuations.
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I. INTRODUCTION

One of the key problems that had to be resolved for the successful realization of III-V nitride semiconductor devices was *p*-type doping. It was first shown by Amano *et al.* that Mg-doped GaN grown by metal-organic chemical-vapor deposition (MOCVD) could be made *p*-type by a postgrowth exposure to low-energy electron beam irradiation.[1] Nakamura *et al.* further demonstrated that thermal annealing of MOCVD GaN:Mg also leads to efficient *p*-type doping.[2] It was later shown that the active hole concentration in highly conductive GaN:Mg can be drastically reduced by an anneal in NH_3 atmosphere or an exposure to atomic hydrogen,[3,4] which points to the critical involvement of hydrogen in the activation of Mg acceptors in GaN.

In principle, there are two microscopic mechanisms that could account for these observations: the formation of Mg-H complexes, which leads to a passivation of the acceptors, or the compensation of the acceptors by hydrogen-induced donors. First-principles total-energy calculations showed that hydrogen can form an interstitial donor in GaN.[5] On the other side, Mg-H complexes, in which the hydrogen atom occupies the antibonding position of one of the nitrogen neighbors, appear to have a more favorable formation energy.[5] The observed increase of the hole mobility upon hydrogenation indicates the formation of such electrically inactive Mg-H complexes.[6] The most decisive support for the existence of Mg-H complexes would, however, be the observation of their local vibrational modes (LVM's). While the first investigation into the LVM's of GaN Mg probably observed H-decorated nitrogen vacancies,[7] modes consistent with the above microscopic model have been reported recently.[8] However, the oscillator strength found for the Mg-H complexes in GaN is considerably smaller than that observed for N-H bonds, e.g., in ZnSe,[9] which sheds some doubt whether the surrounding structure of the majority of Mg atoms has indeed been observed in Ref. 8.

Electronic transport in *p*-type GaN has been studied extensively using the Hall effect and conductivity measurements. However, no information on the complementary thermopower properties of the III-V nitrides is available. Thermopower determines the average energy, with respect to the Fermi level, which is transported by charge carriers under the influence of a thermal gradient[10] and therefore provides alternative information concerning the main conduction path. A temperature difference ΔT across the semiconductor leads to the buildup of a voltage ΔU caused by the majority carriers diffusing from the hot to the cold end. The thermopower or Seebeck coefficient S is given by the ratio $\Delta U/\Delta T$. When the relaxation of the energy E of charge carriers can be described by a relaxation time $\tau\propto E^r$, the Seebeck coefficient for hole conduction is determined from Boltzmann's equation as

$$S=\frac{k}{e}\left[A+\frac{E_F-E_V}{kT}\right], \tag{1}$$

with a constant scattering factor $A=(\frac{5}{2}+r)$, and E_F and E_V denoting the energy of the Fermi level and valence band, as usual. In principle, thermopower measurements allow the determination of the dominant scattering mechanism, of the density of states effective mass, and of the statistical shift of the Fermi level. In addition, the measurement of the sign of the Seebeck voltage provides a definite identification of the type of conductivity even in cases when the concentration of charge carriers is too low for conventional Hall experiments.

Of particular importance for the problem of complex formation vs compensation by hydrogen in GaN:Mg is the fact that in some cases a comparison between thermopower and conductivity experiments has been used to estimate the size of potential fluctuations caused by charged impurities.[11,12] The underlying idea is that charge carriers participating in conductivity are influenced more strongly by mesoscopic potential fluctuations, while for the buildup of a Seebeck voltage under open circuit conditions only activation to the minimum of the fluctuations is necessary. In this case, the activation energy of the thermopower $E_{A,TP}=E_F-E_{V,TP}$ is

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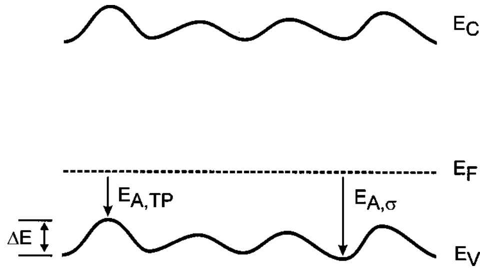

FIG. 1. Schematic representation of the different effective valence-band minima characterizing the transport paths for thermopower and conductivity.

smaller than the respective activation energy $E_{A,\sigma}=E_F-E_{V,\sigma}$ for the conductivity (Fig. 1). The difference $\Delta E=E_{A,\sigma}-E_{A,TP}$ then is a measure of the size of the potential fluctuations. We present here results of comparative Hall effect, conductivity, and thermopower experiments on molecular-beam-epitaxy (MBE) grown GaN:Mg, which in contrast to MOCVD-grown material does not require postgrowth anneal to show *p*-type conductivity. In as-grown GaN:Mg, clear indications for hopping in an impurity band are observed. Upon hydrogenation, $E_{A,\sigma}$ was found to be equal to $E_{A,TP}$ within experimental error. This failure to observe potential fluctuations due to compensating charged impurities provides additional support that neutral Mg-H complexes are formed in GaN.

II. EXPERIMENTAL DETAILS

The thermopower measurements were performed on GaN epitaxial layers grown on *c*-plane sapphire using plasma-induced MBE.[13] The samples were cut into thin stripes (3×9 mm^2) and Au and Al were evaporated as contact material for *p*- and *n*-type material, respectively. The two narrow ends of an individual sample studied were mounted with the sapphire side down onto two copper blocks, whose temperature could be controlled independently by both heating and liquid-nitrogen cooling (Fig. 2). Due to the high thermal conductivity of the sapphire substrate and the comparatively bad thermal contact to the copper blocks, measuring the exact temperature at the contacts used for the determination of the Seebeck voltage is of tantamount importance. This is achieved by Chromel-Alumel thermocouples mounted using silver paste on the metal contacts where they are extending into the sample region between the copper blocks. The Seebeck voltage is then measured between the chromel contacts of each pair using a high impedance voltmeter (Keithley 6517), which allows the accurate determination of the thermopower for sample resistance of up to 200 GΩ. Using this setup, the remaining error in the determination of the Seebeck coefficient is mostly due to a nonperfect knowledge of the sample geometry and can be kept below 10%.

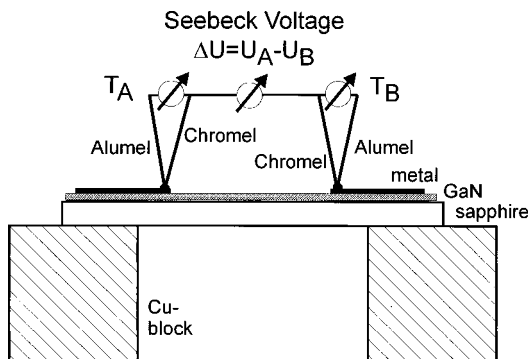

FIG. 2. Experimental setup used to determine the thermopower. The Seebeck voltage is measured between the Chromel elements of two thermocouples.

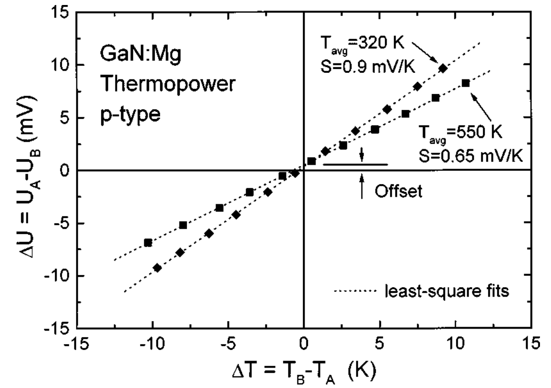

FIG. 3. Determination of the Seebeck coefficient S by variation of the temperature difference ΔT keeping the mean temperature T_{avg} fixed. Using this technique, the influence of contact potentials on the measurement can be minimized.

The simplest way to determine the thermopower is to measure the Seebeck voltage ΔU at a constant temperature difference $\Delta T=T_B-T_A$ of the temperatures T_A and T_B at the two ends of the sample. However, since the two contacts can never be made completely identical and purely Ohmic, an additional contact potential can develop, which distorts the thermopower measurement (offset in Fig. 3). To first order, this effect can be suppressed by measuring ΔU at different ΔT while keeping the main temperature $T_{\text{avg}}=(T_A+T_B)/2$ constant and determining $S(T_{\text{avg}})$ from the slope of $\Delta U(\Delta T)$ using a least-square fit. Figure 3 shows the result of such a measurement of as-grown GaN:Mg. The sign of the Seebeck voltage at the cold end is positive, indicating that indeed the sample is *p*-type. Using this setup, the thermopower properties of GaN could be investigated from $T_{\text{avg}}=110-600$ K.

III. RESULTS

A. *n*-type GaN

As a starting point, we have also investigated the thermopower properties of *n*-type GaN before turning to *p*-type material. Figure 4 shows the temperature dependence of the conductivity and of the thermopower of an unintentionally doped GaN film with an electron concentration $n=2\times10^{18}$ cm^{-3}. At room temperature, the activation energy of the conductivity is $E_{A,\sigma}\approx25$ meV. No clear temperature dependence of the thermopower is observed above 300 K. The extrapolation of the thermopower data to $1/T\rightarrow0$ yields a scattering factor of $A\approx3.8$.

Within the framework of the energy relaxation time approximation, the scattering factor A for scattering by ionized impurities is 4, for scattering by acoustical phonons $A=2$.[10] At high temperatures, scattering in *n*-type GaN is expected to be dominated by the interaction with polar optical phonons.[14]

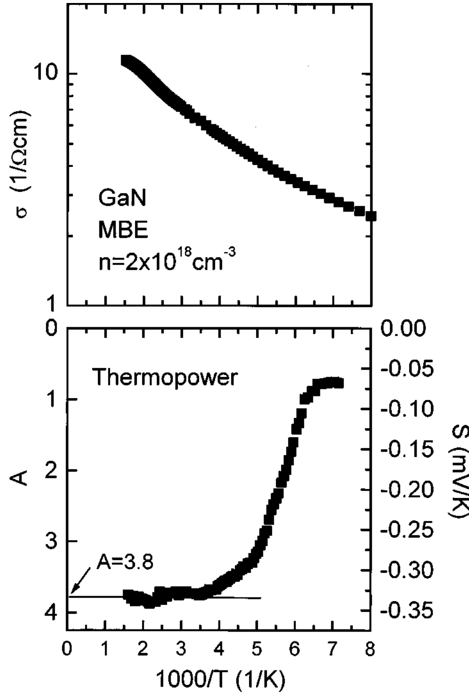

FIG. 4. Temperature dependence of the conductivity σ and of the thermopower S of n-type GaN. Extrapolation of the temperature to $1/T \rightarrow 0$ yields the scattering factor A.

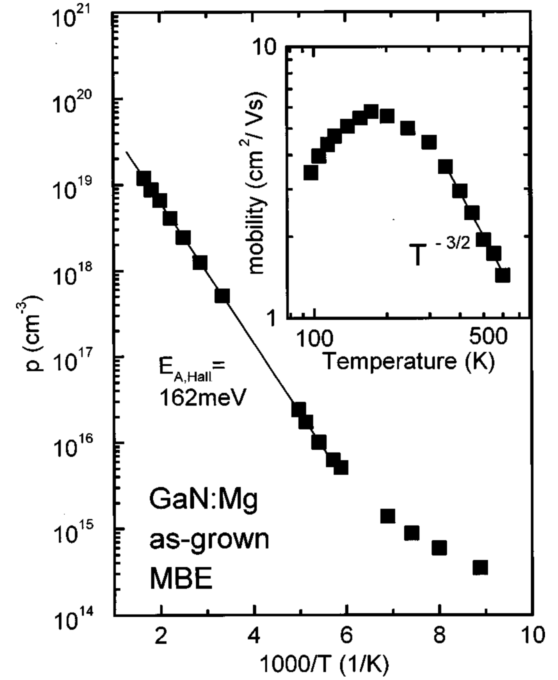

FIG. 5. Arrhenius plot of the effective hole concentration in the Mg-doped GaN sample in the as-grown state. The inset shows the corresponding temperature dependence of the Hall mobility.

The prediction of the corresponding scattering factor is difficult, since this scattering process cannot be easily described by the energy relaxation time approximation. However, $A=3$ has been reported for this scattering mechanism.[15] The observed value of $A \approx 3.8$ lies in between the factors expected for pure polar optical and impurity scattering, respectively. The value close to 4 is in accordance with the relatively low room-temperature mobility of $\mu_n = 110\ \mathrm{cm^2/V\,s}$ exhibited by the sample.

From a comparison of the thermopower and Hall results, the density of states effective mass m^*_{DOS} can be determined according to Johnson and Lark-Horovitz,[16] which provides a valuable cross-check for the quality of the thermopower measurements. In the case of dominant scattering by ionized impurities,

$$S = -\frac{k}{e}\left[\ln\left(1.93\frac{T^{3/2}}{ne}\right) - 3.82 + \frac{3}{2}\ln\left(\frac{m^*_{n,\mathrm{DOS}}}{m_0}\right)\right] \qquad (2)$$

for n-type conductivity, where T is in K, n is in $\mathrm{cm^{-3}}$, and e is in C. Close to room temperature, Eq. (2) provides an estimation of the electron density of states mass in GaN of $m^*_{n,\mathrm{DOS}} = 0.15\ m_0$, close to values for the effective mass m^*_n around 0.23 m_0 determined by other techniques.[17] The marked reduction of the Seebeck coefficient at temperatures below 200 K seen in Fig. 4 is caused by the high donor concentration in the sample investigated. We will discuss such effects in more detail in the context of p-type GaN below.

B. As-grown p-type GaN:Mg

The experiments on p-type GaN have been performed on a GaN:Mg epitaxial layer grown without a buffer layer. Elastic recoil detection analysis determined a Mg concentration of $5\times10^{19}\ \mathrm{cm^{-3}}$ in the layer. Figure 5 shows the results of Hall measurements obtained for the as-grown layer. The activation energy of the hole concentration is $E_{A,\mathrm{Hall}} = 162$ meV, in good agreement with other reports on the Mg acceptor.[18,19] The maximum Hall mobility found in the as-grown sample is $\mu = 6\ \mathrm{cm^2/V\,s}$ at 180 K. At higher temperatures, the mobility decreases as $\mu \propto T^{-3/2}$.

Figure 6 shows the comparison between the temperature dependence of the conductivity and the Seebeck coefficient S for GaN:Mg in the as-grown state. At temperatures between 300 and 500 K, the thermopower has an activation energy $E_{A,TP} = E_F - E_{V,TP} = \approx 130$ meV, slightly larger than the corresponding activation energy $E_{A,\sigma} \approx 110$ meV for the conductivity in this temperature range, and an extrapolated scattering factor of $A \approx 6.5$. At temperatures above 500 K, the activation energy increases to $E_{A,TP} \approx 280$ meV, while the scattering factor becomes $A \approx 3$. At high temperatures, the A therefore approaches the value expected for scattering at polar optical phonons.[14] However, over the whole temperature range, $E_{A,TP}$ is significantly larger than $E_{A,\sigma}$, which is at variance with the thermopower model as discussed in the Introduction. In particular, the possible effects caused by potential fluctuations would lead to $E_{A,TP}$ being smaller than $E_{A,\sigma}$.

It is necessary to include a second transport path to account for the observed thermopower behavior. Hypothetically assuming that electrons from compensating donors would lead to transport in the conduction band, such bipolar transport by electrons and holes would increase the conductivity, but their combined effect would be to decrease the thermopower, in contrast to what is observed here at high temperatures. Until now, we have, however, neglected the contribution of hopping in the acceptor band to the electronic

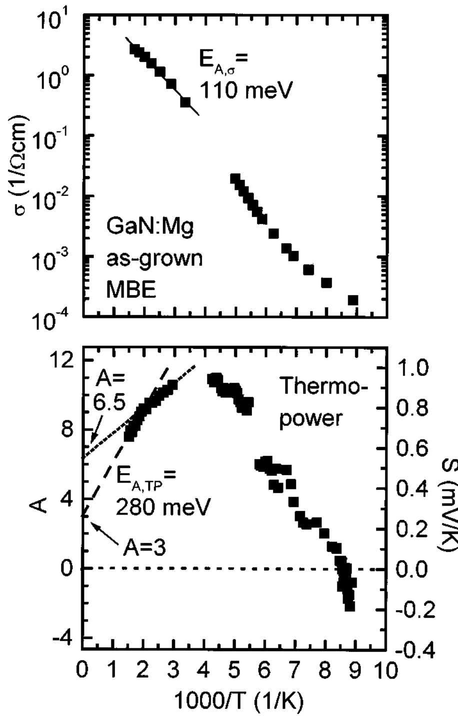

FIG. 6. Temperature dependence of the conductivity σ and of the thermopower S of Mg-doped GaN. S becomes negative below 120 K.

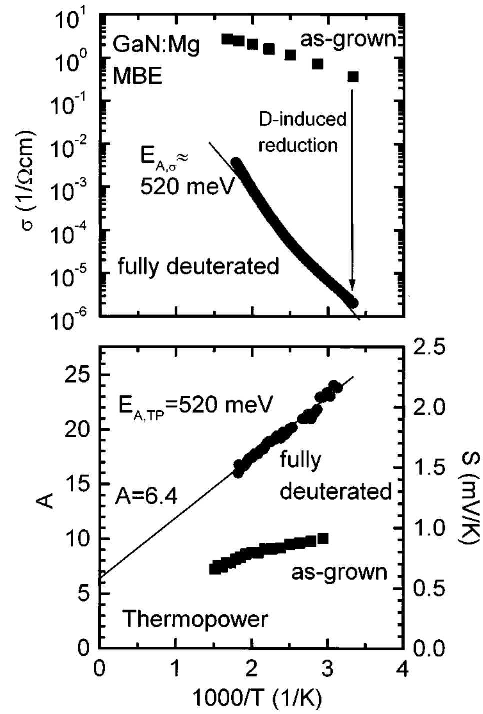

FIG. 7. Comparison of the temperature dependence of the conductivity σ and of the thermopower S of as-grown and fully deuterated Mg-doped GaN. Upon deuteration, the room-temperature conductivity decreases by a factor of 10^5.

transport in GaN:Mg. As can be seen from Fig. 5, only 1% of the charge carriers has been thermally activated to the valence band at room temperature. While the conductivity is dominated by the more mobile holes in the valence band, the thermopower will have comparable contributions from holes in the valence and in the acceptor band. The thermopower resulting from hopping processes has been studied extensively, in particular in disordered systems.[20,21] Due to the relatively deep levels of the dopants, wide band-gap semiconductors provide a unique opportunity to study thermopower under conditions when both transport in extended states as well as hopping in localized states are present.

Convincing evidence for the contribution of hopping in the acceptor band of GaN:Mg to the thermopower comes from the observed reversal of the sign of S at temperatures below 120 K as seen in Fig. 2. A similar behavior has been observed, e.g., by Geballe and Hull in highly doped silicon.[22] Their interpretation was that once the temperature is low enough so that the acceptor band is almost empty of electrons, and provided that the acceptor band does not overlap with the valence band, the impurity band effectively acts as a conduction band and hopping of electrons in the acceptor band gives rise to a Seebeck effect typical for electrons as observed here. Compared to the results obtained for Si,[22] the transition temperature at which the Seebeck coefficient changes sign roughly scales with the depth of the acceptor. In the case of partial compensation, which is very likely for GaN containing residual oxygen donors, the electrons transferred from the donors to the acceptor band additionally contribute to this negative Seebeck coefficient at low temperatures.

The observation of hopping in the Mg-acceptor band clearly indicates that the Fermi level is in or very near to this band. This allows us to resolve an ambiguity that exists in the interpretation of Hall experiments, namely, in the determination of the exact energy of the acceptor level E_A from the activation energy of the Hall concentration $E_{A,\mathrm{Hall}}$. According to standard semiconductor statistics, $E_A = 2E_{A,\mathrm{Hall}}$ when no compensation takes place. In contrast, the presence of compensation leads to $E_A = E_{A,\mathrm{Hall}}$. Although the details of the temperature dependence of the Hall concentration as well as other measurements such as photoluminescence have already indicated the presence of compensating donors in GaN:Mg, the hopping at the Fermi level observed here provides further evidence from transport measurements that the Mg-acceptor level is at $E_A = E_{A,\mathrm{Hall}} \approx 162$ meV.

C. Hydrogenated GaN:Mg

To finally address the influence of hydrogen in GaN:Mg, the samples have been deuterated at 600 °C using a remote dc plasma. The room-temperature conductivity decreased from 4×10^{-1} 1/Ω cm in the as-grown state to 2×10^{-6} 1/Ω cm in the deuterated state. After deuteration, the conductivity was too low to perform Hall experiments. However, thermopower measurements show that the sample remained *p*-type. The results of the high-temperature conductivity and thermopower experiments of deuterated GaN:Mg are shown in Fig. 7. The activation energy $E_{A,TP}$ has significantly increased to 520 meV, as well as the corresponding $E_{A,\sigma}$, which changes from 500 to 540 meV in the temperature range from 300 to 550 K in which the thermopower experiment could be performed. Most notably, the marked difference in the two activation energies observed in the as-grown

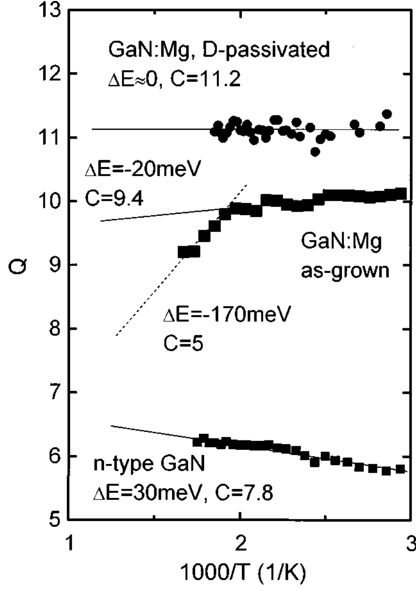

FIG. 8. Temperature dependence of the function Q for n-type GaN, as-grown p-type GaN:Mg, and fully deuterated GaN:Mg.

state has disappeared, i.e., $E_{A,TP} \approx E_{A,\sigma}$ within experimental error. The extrapolation of the thermopower data to $1/T \rightarrow 0$ yields a scattering factor of $A \approx 6$, similar to the value of A obtained for medium temperatures in as-grown GaN:Mg. After hydrogen treatment, thermopower measurements at temperatures significantly below 300 K could not be performed since the sample resistance became too high.

IV. DISCUSSION: THE ROLE OF POTENTIAL FLUCTUATIONS

As discussed in the Introduction, in the presence of potential fluctuations the activation energy of the thermopower $E_{A,TP}$ is expected to be smaller than the activation energy of the conductivity $E_{A,\sigma}$. For p-type conduction, the temperature dependence of the difference $\Delta E = E_{A,\sigma} - E_{A,TP}$ of the activation energies can be expressed using the dimensionless function[23]

$$Q = \ln(\sigma \;\; \Omega \text{ cm}) + \frac{e}{k} S = \ln(\sigma_0 \;\; \Omega \text{ cm}) - \frac{E_F - E_{V,\sigma}}{kT} + A + \frac{E_F - E_{V,TP}}{kT} = C - \frac{\Delta E}{kT}, \tag{3}$$

where σ_0 is the conductivity prefactor. Assuming that A is not temperature dependent, $C = \ln(\sigma_0 \;\; \Omega \text{ cm}) + A$, and $\Delta E = E_{V,TP} - E_{V,\sigma} = E_{A,\sigma} - E_{A,TP}$. When the conductivity is defined independently of the type of charge carrier as $\sigma = |e| n \mu$,

$$Q = \ln(\sigma \;\; \Omega \text{ cm}) - \frac{e}{k} S = \ln(\sigma_0 \;\; \Omega \text{ cm}) - \frac{E_{C,\sigma} - E_F}{kT} + A + \frac{E_{C,TP} - E_F}{kT} = C - \frac{\Delta E}{kT} \tag{4}$$

for n-type conduction with $\Delta E = E_{C,\sigma} - E_{V,TP}$. Extrapolation to $1/T = 0$ therefore provides a combined estimate of A and σ_0.

Figure 8 shows the temperature dependence of Q for all three materials studied here: n-type GaN, as-grown GaN:Mg, and deuterated GaN:Mg. In the case of n-type material, a small positive $\Delta E \approx 30$ meV is found, in accordance with the values of $E_{A,\sigma}$ and $E_{A,TP}$ determined separately. Since the positive value of ΔE suggests the presence of potential fluctuations, we will try to estimate the average amplitude of these fluctuations following an argument by Kane.[24] He finds that for a volume L^3, which contains $N = L^3 N_d$ charged donors of density N_d, the typical statistical deviation $\sqrt{N}$ from this average gives rise to a potential fluctuation

$$\Delta V = \frac{e\sqrt{N}}{4\pi\epsilon\epsilon_0} \frac{1}{L} = \frac{e}{4\pi\epsilon\epsilon_0} (L N_d)^{1/2}. \tag{5}$$

Since the fluctuations are limited by the screening arising from mobile carriers, the relevant length scale L is determined by the Debye screening length $L_{\text{Debye}} = \sqrt{\epsilon\epsilon_0 kT / ne^2}$, where n is the density of mobile charge carriers. Inserting L_{Debye} into Eq. (5),

$$\Delta V = \frac{e}{4\pi\epsilon\epsilon_0} \left[\left(\frac{\epsilon\epsilon_0 kT}{ne^2} \right)^{1/2} N_d \right]^{1/2}. \tag{6}$$

In particular, ΔV is proportional to $N_d^{1/2}$ and $n^{-1/4}$. Starting from a donor concentration of $N_d = 2 \times 10^{18}$ cm^{-3} in the n-type GaN sample studies, assuming complete activation of donors, i.e., $n = N_d$, and using $\epsilon = 8.5$, a value of $e\Delta V \approx 12$ meV is found at room temperature. Due to the weak $T^{1/4}$ dependence of ΔV and the limited temperature range investigated in Fig. 8, we neglect the temperature dependence of ΔV in the remainder of the discussion. Taking into account the assumptions used in the derivation of Eq. (6), we find a good agreement of $e\Delta V$ and ΔE for n-type GaN, which adds further confidence that our assignment of a positive ΔE to potential fluctuations is indeed correct.

As discussed above, the thermopower properties of the as-grown GaN:Mg material studied are significantly influenced by hopping in the acceptor band, and cannot be interpreted in terms of potential fluctuations, as evidenced by the negative values of ΔE. Using the results of the Hall effect measurements of the as-grown p-type material (carrier concentration p and ionized acceptor concentration N_a at room temperature of $p = N_a = 5 \times 10^{17}$ cm^{-3}), fluctuations of the order of only $e\Delta V \approx 8$ meV would be expected.

We will now estimate the potential fluctuations in the hydrogen-treated sample. Since Hall measurements could not be performed after complete hydrogenation, we determine the hole concentration p from the change in conductivity shown in Fig. 7. Under the assumption that the mobility is unchanged, we obtain $p \approx 10^{13}$ cm^{-3}. In the case of passivation by complex formation, $p = N_a$, and the size of the potential fluctuations will be of the order of a few meV. In contrast, assuming that compensation takes place upon hydrogenation of GaN:Mg, $p \ll N_a = 5 \times 10^{17}$ cm^{-3} and N_d. In this case, we find $e\Delta V \approx 140$ meV, since screening by free charge carriers is markedly reduced. Even when changes in the mobility are taken into account for the estimation of p

from the conductivity measurements, potential fluctuations of the order 100 meV are expected in compensated GaN:Mg. However, within experimental accuracy $\Delta E \approx 0$ in fully deuterated GaN Mg ($|\Delta E| < 10$ meV). We conclude that in this material no potential fluctuations due to charged impurities (Mg acceptors and hydrogen-related donors) exist. Our data therefore are in favor of electrically inactive Mg-H complexes in GaN:Mg rather than compensating H-related donors. Should hydrogenation lead to the formation of nitrogen vacancies (V_N), Mg compensation by V_N can similarly be ruled out in favor of the formation of Mg-V_N complexes.

V. SUMMARY AND CONCLUSIONS

We have investigated the transport properties of *n*- and *p*-type GaN:Mg using the complementary methods of thermopower, Hall effect, and conductivity. In particular, it was shown that in MBE-grown Mg-doped material, the thermopower is due to transport by holes in the valence band as well as hopping in the acceptor band, as seen most notably by the change of the sign of the Seebeck effect at low temperatures. However, the large discrepancy of the activation energies of the thermopower and the conductivity at high temperatures clearly indicates that conduction in the valence band alone cannot adequately describe electronic transport in this material, but that hopping in the acceptor band is also relevant at device operating temperatures. The same should hold for the participation of deep acceptor or donor states in the conduction processes in other wide band-gap semiconductors such as II-VI compounds or diamond. Upon introduction of deuterium into Mg-doped GaN, the activation of the thermopower and of the conductivity are identical, which excludes the presence of pronounced potential fluctuations caused by compensation of Mg acceptors and provides additional evidence that electrically inactive Mg-H complexes are formed in GaN.

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