Low temperature electron mobility exceeding $10^4$ cm$^2$/V s in MOCVD grown $\beta$-Ga$_2$O$_3$

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ABSTRACT

We report on record electron mobility values measured in lightly Si doped homoepitaxial $\beta$-Ga$_2$O$_3$ grown by metal-organic chemical vapor deposition. The transport properties of the films were studied using temperature-dependent Hall measurements. Numerous (010) $\beta$-Ga$_2$O$_3$ layers grown at different conditions showed peak electron mobility exceeding $10^4$ cm$^2$/V s at low temperature (LT), with the highest value of 11 704 cm$^2$/V s at 46 K. The room temperature electron mobilities of the films were between 125 cm$^2$/V s and 160 cm$^2$/V s with the net background charge concentration between $\sim 5 \times 10^{15}$ cm$^{-3}$ and $\sim 2 \times 10^{16}$ cm$^{-3}$. The obtained LT mobility values for $\beta$-Ga$_2$O$_3$ were found to be comparable to or higher than the highest LT electron mobilities in bulk SiC and GaN films in the literature. The results demonstrate the capability of metalorganic chemical vapor deposition (MOCVD) for growing high quality ultrapure $\beta$-Ga$_2$O$_3$ epitaxial films that are suitable for high power electronic device applications.

$\beta$-Ga$_2$O$_3$ has attracted broad interest for power electronic applications owing to its superior material properties, including a large bandgap energy of $\sim 4.9$ eV, a high critical breakdown field of $\sim 8$ MV/cm, and the availability of freestanding high quality $\beta$-Ga$_2$O$_3$ substrates grown directly from the melt. The high critical breakdown field ($E_c$) leads to a large Baliga’s figure of merit (BFOM = $\varepsilon\mu E_c^2$, where $\varepsilon$ is the dielectric constant and $\mu$ is the carrier mobility) of $\sim 3400$, which is four to ten times higher than that of GaN and SiC. Moreover, the controllable $n$-type doping and compensating acceptor doping achievable using various growth methods provide a suitable platform for realizing $\beta$-Ga$_2$O$_3$ based power devices. Its application for power electronics has already been demonstrated in key power devices, including MESFETs, Schottky barrier diodes (SBDs), and MOSFETs. To achieve the optimal performance of these devices, including high breakdown voltage and low on-resistance, realizing $\beta$-Ga$_2$O$_3$ drift regions with high electron drift mobility and low charge concentration is essential. This necessitates high quality epitaxial films with low dislocation density and defect density, as well as low background or minimal unintentional impurity concentration in the film.

Recently, the first high electron mobility of $\sim 2800$ cm$^2$/V s at low temperature (LT) was demonstrated in a two-dimensional electron gas (2DEG) developed by molecular beam epitaxy (MBE) using the modulation-doped $\beta$-(Al,Ga)$_2$O$_3$/$\beta$-Ga$_2$O$_3$ heterostructure. The high 2DEG mobility was attributed to better screening of the scattering centers and reduced impurity scattering due to the spatial separation between the dopant impurities and the 2DEG, but the mobility calculations indicated the existence of $\sim 1 \times 10^{12}$ cm$^{-3}$ background impurities in the UID Ga$_2$O$_3$ layer. Following this, subsequent improvement in the material quality of bulk homoepitaxial $\beta$-Ga$_2$O$_3$ films grown by metalorganic chemical vapor deposition (MOCVD) and hydride vapor phase epitaxy (HVPE) produced films with enhanced LT electron mobilities ranging between $\sim 3500$ cm$^2$/V s and $\sim 5000$ cm$^2$/V s. These improvements are primarily due to the reduced total background impurity concentrations to the low $10^{16}$ cm$^{-3}$ range, which are unintentionally incorporated into the $\beta$-Ga$_2$O$_3$ films. Theoretical calculations show
that for a clean $\beta$-Ga$_2$O$_3$ sample with no compensation acceptors ($N_a = 0$) and low donor concentration ($N_d \sim 1 \times 10^{15}$ cm$^{-3}$), an electron mobility of $>10^4$ cm$^2$/V s can be achieved at low temperatures. Similarly, in our recent work, where we demonstrated a RT mobility of 176 cm$^2$/V s with a free charge density of $7.4 \times 10^{14}$ cm$^{-3}$ and a peak mobility of $\sim 3500$ cm$^2$/V s at 54 K, we predicted the possibility of achieving an LT mobility of $10^4$ cm$^2$/V s by growing clean $\beta$-Ga$_2$O$_3$ films with the shallow donor concentration of $5 \times 10^{15}$ cm$^{-3}$ and the compensation acceptor density of $\sim 1 \times 10^{13}$ cm$^{-3}$.

In this letter, we present our recent experimental data that demonstrate the growth of superclean $\beta$-Ga$_2$O$_3$ epitaxial films with electron mobility over $10^4$ cm$^2$/V s by MOCVD. The highest LT electron mobility obtained in this work is $11 \, 704$ cm$^2$/V s, which is the highest value ever reported for $\beta$-Ga$_2$O$_3$, exceeding the theoretically predicted value for a clean $\beta$-Ga$_2$O$_3$. The obtained results are comparable with the highest LT mobility value of $12 \, 400$ cm$^2$/V s reported for UID 4H-SiC films and higher than the highest LT mobility value of $7386$ cm$^2$/V s obtained in bulk GaN grown by hydride vapor phase epitaxy.

In this work, four $\sim 2.5$ to 3.2 $\mu$m thick (sample Nos. 1–4) lightly Si doped $\beta$-Ga$_2$O$_3$ thin films were grown on Fe-doped (010) $\beta$-Ga$_2$O$_3$ substrates (Novel Crystal Technology) using Agnitron Technology’s Agilis R&D MOCVD system. Triethylgallium (TEGa) and pure oxygen (5N) were used as a precursor for Ga and $O_2$, respectively. Ar (6N) gas was used as a carrier to deliver the TEGa vapor into the reactor. The Ar and $O_2$ gases were passed through point-of-use purifiers to reduce the impurity to below the parts per million level. The layers were grown at $\sim 5$ $\mu$mol/h using growth pressure, substrate temperature, and $O_2$/TEGa ratio in the ranges of 50–150 Torr, 800–1000 $^\circ$C, and 1000–2500, respectively. The films’ thicknesses were measured using Filmetrics by growing the films on sapphire substrates that were simultaneously loaded into the reactor along with the $\beta$-Ga$_2$O$_3$ substrates. Silicon was introduced into the films using silane (SiH$_4$) diluted in helium precursor. For each of the four films, the molar flow rate of silane was held constant to $2.5 \times 10^{-12}$ mol/min.

The surface morphologies and crystal qualities of the films were analyzed by atomic force microscopy (AFM) and high-resolution x-ray diffraction (HRXRD). The results have shown comparable crystal and surface qualities for each film, and here, we present representative data for one of the films. Figure 1(a) shows the 2D AFM image of the representative film (sample No. 3) measured from a 20 $\times$ 20 $\mu$m$^2$ scan area. The surface showed elongated grooves along the [001] crystal orientation and an rms roughness of $\sim 1.4$ nm. The observed features are characteristics of step-bunching growth, which was previously observed for the films grown by MBE and MOCVD. In Fig. 1(b), the HRXRD (020) peak of the same epitaxial layer is shown. The XRD full width at half maximum (FWHM) of the (020) peak from a 29-ω scan is $\sim 43$ arc sec, which is comparable to the literature values reported for epitaxial films and bulk $\beta$-Ga$_2$O$_3$ substrates.

The electronic properties of the samples were characterized using temperature-dependent Hall measurement in van der Pauw configuration. Ohmic contact pads with a metal stack of Ti/Pt/Au (50 nm/20 nm/150 nm, Fig. 2(a)) were deposited by electron beam evaporation at the corners of the rectangular samples. This was followed by a rapid thermal annealing at 500 $^\circ$C in a N$_2$ environment. The contacts remained Ohmic in the entire measurement temperature range from RT to 45 K as confirmed by the current-voltage (IV) characteristics of the contacts. Representative current-voltage (IV) characteristics of the metal contacts of sample No. 2 measured at RT and 45 K are shown in Figs. 2(b) and 2(c), respectively. This Ohmic behavior ensured the reliability of the Hall measurements, especially in the cryogenic temperature range.

Figures 3(a) and 3(b) present the temperature-dependent Hall mobility and sheet resistance of the films, respectively. The measured LT electron mobility for the films ranges between 125 cm$^2$/V s and 160 cm$^2$/V s with the corresponding free carrier concentration between $5 \times 10^{13} 2 \times 10^{16}$ cm$^{-3}$ (Table I), which are comparable to RT electron mobilities previously obtained using MOCVD. As the temperature reduces, the electron mobility increases, showing peak LT values of 7750 cm$^2$/V s, 10 194 cm$^2$/V s, 11 211 cm$^2$/V s, and 11 704 cm$^2$/V s, for sample Nos. 1–4, respectively. These peak mobilities are substantially higher than the recently reported LT electron mobilities of 4984 cm$^2$/V s and 5000 cm$^2$/V s for MOCVD and HVPE grown homoepitaxial Si doped Ga$_2$O$_3$ films, suggesting further improvement in the materials quality obtained in this work.

Table I compares the RT and peak (LT) electron mobilities measured from the epitaxial layers grown in the current work with the previously reported state-of-the-art values both for the epitaxial films grown by various methods and bulk substrates. As shown in Table I, sample Nos. 1–4 have comparable RT electron mobilities, but higher LT electron mobilities than the previously reported results. For a pure or lightly doped $\beta$-Ga$_2$O$_3$ material, the
RT electron mobility is dominantly limited by the long-range interaction between electrons and polar optical phonons.\textsuperscript{21,25,38} The relatively lower RT mobility values below 150 cm\textsuperscript{2}/V s for sample No. 1 and 4 are attributed to the existence of a low mobility parasitic channel near the substrate surface. The frequently observed unintentional but heavy accumulation of donor impurities at the film/substrate interface (e.g., Si with up to $1 \times 10^{19}$ cm\textsuperscript{-3} concentration as measured by SIMS)\textsuperscript{32,39} and surface riding of Fe into the epilayers from the substrate\textsuperscript{40} could contribute to this low mobility channel near the substrate interface. Better surface treatment or introducing a semi-insulating layer at the beginning of the growth should eliminate the parasitic channel and further improve the RT mobility close to the theoretically predicted value of $\sim$200 cm\textsuperscript{2}/V s.\textsuperscript{10,11,25,35}

As the temperature decreases, the scattering from ionized impurities becomes increasingly important in controlling the LT electron mobility.\textsuperscript{25,37} The peak electron mobilities measured for sample Nos. 2–4 are more than $10^4$ cm\textsuperscript{2}/V s, surpassing the theoretically predicted values for an ideal sample with a donor concentration of $<1 \times 10^{19}$ cm\textsuperscript{-3} and no compensating acceptors in the material.\textsuperscript{25} This suggests that the lightly doped MOCVD grown $\beta$-Ga\textsubscript{2}O\textsubscript{3} films in the current work are remarkably pure.

The mobility values obtained here are also compared to the highest LT electron mobilities for bulk UID SiC and GaN films reported in the literature.\textsuperscript{4,25,41} As summarized in the table, the results obtained for our films are comparable to the peak values reported for SiC grown by atmospheric pressure chemical vapor deposition (APCVD)\textsuperscript{26} but much higher than the LT electron mobility reported for a freestanding GaN grown by hydride vapor phase epitaxy (HVPE).\textsuperscript{27} This suggests that the material purity of epitaxial $\beta$-Ga\textsubscript{2}O\textsubscript{3} films realized using MOCVD growth is comparable to or higher than what is achievable for the state-of-the-art SiC and GaN materials. Therefore, in addition to the excellent fundamental material properties of $\beta$-Ga\textsubscript{2}O\textsubscript{3},\textsuperscript{12} the capability of the growth of high quality films indicates the superiority of $\beta$-Ga\textsubscript{2}O\textsubscript{3} for power electronics applications over its competing wide bandgap semiconductors, such as SiC and GaN. With such bulk film purity, the 2DEG at the $\beta$-(Al, Ga)\textsubscript{2}O\textsubscript{3}/$\beta$-Ga\textsubscript{2}O\textsubscript{3} heterostructure interface could potentially reach much higher RT and LT electron mobilities since the quantum confinement at the heterojunction enables better screening of both the phonon and impurity scattering centers.\textsuperscript{19,42,43}

In Fig. 3(b), the temperature-dependent sheet resistance curves for the same samples are presented. At room temperature, the sheet resistance of the films is between $\sim$10 kΩ/□ and $\sim$35 kΩ/□. The resistance values then decrease with temperature, following the increasing trend of electron mobility, until they reach the minimum values ranging between 2.7 kΩ/□ and 4.7 kΩ/□ at $\sim$93 K. At lower temperatures below 93 K, the sheet resistance increased exponentially due to the freezeout of bulk carriers.\textsuperscript{44}
The measured Hall charge density as a function of the inverse temperature (1000/T) is shown in Fig. 4. At RT, Hall carrier densities ranging between ~5 × 10^{15} cm^{-3} and ~2 × 10^{16} cm^{-3} were measured for the samples (Table I). To estimate the background impurity concentration and the compensation level in the β-Ga2O3 layers, the temperature dependence of the Hall charge density was fit using the charge neutrality equation. Based on our analysis, the best fit to the experimental data was obtained by including three donor states and a compensating acceptor, with the charge neutrality equation expressed as

\[ n + N_a = \sum_{i=1}^{3} \frac{N_{di}}{1 + 2 \exp \left( \frac{E_c - E_{di}}{k_B T} \right)} \]

where \( n \) is the free electron density and \( N_a \) is the concentration of compensating acceptors. \( N_{d1}, N_{d2}, \) and \( N_{d3} \) represent concentrations of the three donors corresponding to donor energies of \( E_{d1}, E_{d2}, \) and \( E_{d3} \), respectively. \( E_F \) is the Fermi level at the measurement temperature \( T \), which can be estimated by \( n = N_c e^{(E_c - E_F)/k_B T} \), where \( E_c \) is the energy of the conduction band edge, \( N_c \) is the effective density of state in the conduction band, and \( k_B \) is the Boltzmann constant. Here, \( N_c \) is estimated analytically using an electron effective mass of \( m^* = 0.313m_0 \), but \( N_{d1}, N_{d2}, N_{d3}, E_{d1}, E_{d2}, E_{d3} \), and \( N_a \) were free parameters extracted from the best fit, as shown in Fig. 4.

The values of the extracted parameters for the four samples are summarized in Table II. For each sample, the three donor levels are identified at activation energies of \( E_{d1} = 35.2 \text{ meV}, E_{d2} = 60 \text{ meV}, \) and \( E_{d3} = 120–140 \text{ meV} \) below the conduction band edge with the corresponding donor concentrations for each sample presented in the table. Temperature-dependent charge density in previous studies was well fitted by a two donor model in which two donor activation energies were extracted. The source of the shallow donor level (i.e., \( E_{d1} \approx 20 \) to 35 meV) was attributed in every case to the contribution from Si incorporated into the material either intentionally or unintentionally during the material growth. For the second donor activation energy, values of 60 meV, 80 meV, 110 meV, and 120 meV were reported at different times. The origin of these donors has not yet been conclusively identified, but possible sources, including antisites, interstitials, and extrinsic impurities such as Si occupying the octahedrally coordinated Ga\((a)\) site, were suggested. In our samples, the best fit was obtained using the three donor model, where the extracted shallow donor located

![FIG. 4. Hall carrier density (Log scale) as a function of inverse temperature (1000/T) for the four MOCVD grown samples. The symbols are the measured data, and the solid lines present the fitting of the charge density calculated from the charge neutrality equation using the three-donor model. Room temperature free carrier densities are 2.2 × 10^{16} cm^{-3} (No. 1), 8.7 × 10^{15} cm^{-3} (No. 2), 5.3 × 10^{15} cm^{-3} (No. 3), and 1.5 × 10^{16} cm^{-3} (No. 4). The extracted donor and acceptor concentrations and donor levels are summarized in Table II.](image-url)
at $E_{d1} = 35.2$ meV is attributed to Si impurity, but the origin of the second and third higher activation energy donors still needs further investigation.

The third donor level extracted from the fittings for the samples studied in this work could potentially originate from a parasitic channel near the substrate surface. This low mobility parasitic channel could adversely impact the measured RT electron mobility and lead to slightly lower RT mobility values as compared to the previous reports, as shown in Table I. The compensating acceptor concentrations extracted for sample Nos. 2–4 are $8.0 \times 10^{14}$ cm$^{-3}$, $8.0 \times 10^{14}$ cm$^{-3}$, and $7.0 \times 10^{14}$ cm$^{-3}$, respectively, while it is difficult to get reliable fitting of $N_a$ for sample No. 1, suggesting even lower acceptor concentration. The fraction of donor compensation by the acceptors is estimated using $N_a \times 100\%/(\Sigma_{i=1,2} N_{di}$, providing, correspondingly, the compensation levels of 7.5%, 10.5% and 3.7% for the samples. The low compensation charge density along with the lightly doped layers is the basis for the high electron mobility at low temperatures, which is crucial for developing high performance power electronics. Nevertheless, detailed characterizations such as deep level transient spectroscopy (DLTS) may be helpful, but are beyond the scope of this work, to understand the origin of the higher activation energy donors. If the origins of these donors are identified and understood so that they can be minimized, then MOCVD grown $\beta$-Ga$_2$O$_3$ epitaxial films with much higher electron mobilities could be feasible.

In summary, high purity lightly Si doped homoepitaxial $\beta$-Ga$_2$O$_3$ films with good surface morphology and crystalline quality were realized by MOCVD. The transport properties of the films were studied using temperature-dependent Hall measurement, exhibiting peak electron mobilities consistently exceeding $10^5$ cm$^2$/V s for multiple samples. In particular, one of the epitaxial films showed a peak electron mobility of $11.704$ cm$^2$/V s at 46 K, setting a record for $\beta$-Ga$_2$O$_3$. The samples were found to exhibit RT electron mobilities ranging between 125 cm$^2$/V s and 160 cm$^2$/V s with the corresponding free carrier density between $\sim5 \times 10^{15}$ and $2 \times 10^{16}$ cm$^{-3}$. We postulate the existence of a parasitic channel near the substrate surface, which could have lowered the RT mobility while inactive at low temperatures due to carrier freezeout. The mobility values obtained for $\beta$-Ga$_2$O$_3$ were found to be comparable to or higher than the highest LT electron mobilities reported for SiC and GaN in the literature. The results presented here demonstrate the capability of MOCVD for growing high quality ultrapure epitaxial $\beta$-Ga$_2$O$_3$ films required for its practical use in high-performance power device electronics.

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**TABLE II.** Extracted parameters from fitting the temperature dependence of the Hall charge density using the charge neutrality equation.

| Samples | $E_{d1}$ (meV) | $N_{d1}$ (cm$^{-3}$) | $E_{d2}$ (meV) | $N_{d2}$ (cm$^{-3}$) | $E_{d3}$ (meV) | $N_{d3}$ (cm$^{-3}$) | $N_a$ (cm$^{-3}$) |
|---------|----------|----------------|----------|----------------|----------|----------------|----------------|
| No. 1   | 35.2     | $5.0 \times 10^{15}$ | 60.0     | $1.2 \times 10^{16}$ | 120.0    | $1.6 \times 10^{16}$ | ...   |
| No. 2   | 35.2     | $4.6 \times 10^{15}$ | 60.0     | $2.5 \times 10^{15}$ | 120.0    | $3.5 \times 10^{15}$ | $8.0 \times 10^{14}$ |
| No. 3   | 35.2     | $2.6 \times 10^{15}$ | 60.0     | $2.0 \times 10^{15}$ | 140.0    | $3.0 \times 10^{15}$ | $8.0 \times 10^{14}$ |
| No. 4   | 35.2     | $4.7 \times 10^{15}$ | 60.0     | $7.5 \times 10^{15}$ | 120.0    | $6.5 \times 10^{15}$ | $7.0 \times 10^{14}$ |
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