



Article Toward Precise n-Type Doping Control in MOVPE-Grown β-Ga₂O₃ Thin Films by Deep-Learning Approach

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Abstract: In this work, we train a hybrid deep-learning model (fDNN, Forest Deep Neural Network) to predict the doping level measured from the Hall Effect measurement at room temperature and to investigate the doping behavior of Si dopant in both (100) and (010) β -Ga₂O₃ thin film grown by the metalorganic vapor phase epitaxy (MOVPE). The model reveals that a hidden parameter, the Si supplied per nm (mol/nm), has a dominant influence on the doping process compared with other process parameters. An empirical relation is concluded from this model to estimate the doping level of the grown film with the Si supplied per nm (mol/nm) as the primary variable for both (100) and (010) β -Ga₂O₃ thin film. The outcome of the work indicates the similarity between the doping behavior of (100) and (010) β -Ga₂O₃ thin film via MOVPE and the generality of the results to different deposition systems.

Keywords: β-Ga₂O₃; deep learning; doping; MOVPE

1. Introduction

Recently, β -Ga₂O₃ has attracted significant attention in academia and industry as one of the potential candidates for power-electronics application due to its wide bandgap of 4.9 eV and a high theoretical breakdown voltage of up to 8 MV/cm² [1]. A critical advantage of β -Ga₂O₃ is the availability of large, high-quality native substrates grown from the melt using various techniques, such as the Czochralski method [2,3], the floatingzone techniques [4,5], edge-defined film-fed growth (EFG) [6], and the vertical Bridgman method [7], which allows for the economically practical manufacturing of β -Ga₂O₃ when compared to other wide-bandgap semiconductors. Due to these properties, β -Ga₂O₃ is considered a potential alternative to GaN and SiC for future power electronics [8]. As for most semiconductor device applications, a well-controlled doping process is crucial, including a precise doping level, doping uniformity, and doping profile. Group IV elements, such as Ge [9,10], Sn [11], and Si [12], are the potential n-type dopants to the β -Ga₂O₃ thin films. Si, due to a wide doping range (1 × 10¹⁷ to 8 × 10¹⁹ cm⁻³) and low "memory effect" in the reactor chamber [12], has attracted great interest for the doping of β -Ga₂O₃ thin film grown by MOVPE.

Doping is a kinetically controlled process that is strongly influenced by growth conditions. However, the nonlinear correlation between the parameters in the MOVPE process (i.e., the metal and oxidant precursor concentrations, temperature, dopant concentration, etc.) creates a multidimensional parameter space such that optimization for a specific application is challenging and time-consuming. Therefore, finding a predictive empirical relation that results in the desired doping level has become a challenge in thin-film research. In the field of MOVPE-grown β -Ga₂O₃, the observed doping level is usually limited to two situations: (1) only the relation between the doping level and a single process parameter is



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). revealed, e.g., the chamber pressure [13,14] and the concentration of the Si precursor [12,15], and (2) the reported results are usually collected in different deposition systems, which are difficult to be applied to other systems directly. To deal with the above limitations, datadriven approaches such as machine learning and deep learning are seen as promising tools that are applied to explore the ample process-parameter space and understand the nonlinear relationship between them [16], and have already demonstrated a wide application in different research topics such as bulk crystal growth [17–20], thin-film growth [21–23], molecular-property prediction [24–26], and chemical-reaction development [27]. Nevertheless, the available dataset in lab-level research is usually small due to the limited workforce and faculty resources, which is a critical issue for a data-driven methodology such as deep learning. Building a predictive model for the lab-level research is challenging since the number of fabricated samples (n) is usually smaller than the number of process parameters (p), which is commonly known as the "large p, small n" problem [28], and raises both the risk of overfitting and the challenge of high-variance gradients while training the neural network. In the recent progress of the field of bioinformatics, Kong et al. [29] proposed a novel deep-learning framework to vastly enhance the prediction accuracy with a hybrid structure of the random forest and neural network, named Forest Deep Neural Network (fDNN). This framework has a relatively higher prediction accuracy than existing methods for the "large p, small n" problem. Inspired by their work, we implemented the fDNN model to investigate the doping behavior of β -Ga₂O₃ thin film. The parameter space of our dataset was chosen to cover several orders of magnitude in order to generalize the modeling results over the space and to identify the parameter windows for different doping levels. The results of the fDNN were used to explain the experimental findings within the multidimensional parameter space, and an empirical relation was provided based on the insight of the fDNN. In the current work, we identified the critical growth parameter describing the free-electron concentration of the MOVPE-grown Si-doped β -Ga₂O₃ thin film (both (100) and (010) orientations) with the help of the fDNN model and our experimental dataset. The dataset used for the model training consisted of 104 samples of (100)-oriented films with a low defect density [30,31], and all of the incorporated Si in the films was expected to be electrically active. To further examine the robustness of the developed model, another dataset consisted of 15 samples of (010)-oriented films from our previous results [12], and results from the literature were extracted and fitted with satisfying results [15].

2. Materials and Methods

2.1. Experimental

The MOVPE system used in this work consisted of a vertical showerhead low-pressure reactor (Structured Materials Industries, Inc., Piscataway Township, NJ, USA) equipped with a rotating susceptor that was utilized for the deposition of n-type β -Ga₂O₃ thin films. Triethylgallium (TEGa) was used as the metalorganic precursor for Ga and Tetraethylorthosilicate (TEOS) was used as the metalorganic precursor for the n-type doping by Si, while O₂ (5N) was used as the oxidant. High-purity Ar (5N) acted as the push gas. High-quality, semi-insulating (Mg-doped, [Mg] = $\sim 2 \times 10^{18}$ cm⁻³) (100) β -Ga₂O₃ substrates were grown by the Czochralski method [2] at Leibniz-Institut für Kristallzüchtung (IKZ) and commercial, semi-insulating (Fe-doped, [Fe] is in the range of 1 to 4×10^{18} cm⁻³) (010) β -Ga₂O₃ substrates were grown by the EFG method [6]. To reduce possible Si contaminants on the substrate surface, the substrates were immersed in hydrofluoric acid (5%) for 5 min and then rinsed with deionized water before being loaded into the chamber. The experimental parameter space for the β -Ga₂O₃ thin film dataset in this work is summarized in Table 1. The electrical data (doping level and mobility) were obtained by conductivity and Hall Effect measurements at room temperature in a van-der-Pauw configuration using small InGa eutectic contacts that exhibited ohmic behavior. The chemical concentration of the incorporated Si dopant and the thickness of the grown films were determined by the secondary ion mass spectrum (SIMS) performed by RTG Mikroanalyse GmbH Berlin. The

thickness of the grown films was also determined by a LayTec in situ process monitoring system (multi-wavelength reflectance, EpiTT and EpiNet 2017). Since the reflectance spectroscopy required a significant contrast in the refractive index between film and substrate, we used reference films that were simultaneously grown in the same runs on sapphire substrates. For several deposition runs of β -Ga₂O₃ on (100) and (010) β -Ga₂O₃ substrate, we could verify the same or comparable thickness by transmission electron microscopy (TEM) and SIMS to that of the films deposited on sapphire substrates.

Table 1. The process parameters and their range in this work.

Parameter	Range
Growth temperature	700–825 °C
Chamber pressure	5–70 mbar
TEGa molar flow	$3 imes 10^{-6}$ – $3 imes 10^{-5}$ mol/min
TEOS molar flow	$1 imes 10^{-11}$ – $3 imes 10^{-8}$ mol/min
Oxygen molar flow	3×10^{-5} – 5×10^{-2} mol/min
Push gas	1800–4000 sccm

2.2. Machine-Learning Methodology

The fDNN model consists of two parts: A random-forest part and a neural-network part, as shown in Figure 1. The random-forest part is a feature selector to learn sparse feature representations from raw inputs under the supervision of training labels, and the neural network part is to predict outcomes for the new feature representations selected by the random-forest part. The detailed derivations of the random forest and the Forest Deep Neural Network refer to Leo Breiman [32] and Kong et al. [29].



Figure 1. The simplified structure of the fDNN model in this work based on Ref. [29].

The experimental data that were input into the machine-learning model can be categorized into two groups: growth parameters such as precursor concentration, chamber pressure, growth temperature, etc., and measurement values such as film growth rate and free-carrier concentration. The process parameters were controllable and set by the operator, and the measurement values were collected by the methodologies described in the experimental section. The following 6 inputs (growth parameters) were entered for the initiation training: growth temperature (°C), chamber pressure (mbar), TEGa molar flow (mol/min), TEOS molar flow(mol/min), oxygen molar flow(mol/min), and push gas (sccm). The target variable was the free-carrier concentration (doping level) that was measured by the Hall Effect measurement. In general, the free-electron concentration measured by the Hall Effect measurement may deviate from the chemical-dopant concentration due to structural defects such as incoherent twin boundaries [33], the point defects such as gallium vacancies [1,34], or the residual impurities such as Mg [35] and Fe [36], which significantly lower the doping efficiency and deteriorate the electrical properties. For the films investigated in this work, the structural defects were not observed under transmission electron microscopy (TEM) [12] or eliminated by applying substrate miscut [30,37] for the (010) and (100) cases, respectively, and the residual impurities in the selected samples of this work were also far below the doping level. Therefore, the point defects became the primary compensating acceptor in our consideration. The gallium vacancies and their complexes might play a dominant role [38,39]. In some related works [12,40], the concentration of the gallium vacancy was found with to be $<1 \times 10^{16}$ cm⁻³, which is far lower than the dopant concentration range (10^{17} – 10^{19} cm⁻³) that we investigated in this work. As a result, the measured free-electron concentration was expected to be close to the chemical concentration of Si measured by SIMS.

Subsequently, our dataset was randomly subdivided into a training set and a testing set, with 85% of the data being randomly selected into the training set and the remaining 15% sample being classified into the testing set by default. The performances of the model were expressed by statistical regression metrics: coefficient of determination (R²) and root mean square error (RMSE). This work applied a k-fold cross-validation method to evaluate the trained model [41], where k was 10. The method partitioned the training dataset into k non-overlapping sets, and a total of k models were fit and evaluated.

The training of the fDNN model consisted of two steps. First, the training data, including labels, were used to fit the random-forest model, and second, the predictions that were transformed by one-hot encoding from each decision tree for all sets of the process parameters were fed into the neural network. In the current implementation, the input for the neural network was an $n \times M \times 2$ tensor rather than an $n \times M$ matrix, where n is the input number of process parameter sets, and M is the number of trees in the random forest. In the present model, M was chosen as 65, which is in the suggested range from Ref. [41], and there was no significant improvement by further increasing M during the model training. For the second model training step, the number of layers and the neuron number of a hidden layer were determined by 10-fold cross-validations in the range of 1–3 hidden layers and 9–16 neurons per hidden layer. The results are given in Supplementary Materials Table S1. The highest \mathbb{R}^2 , 0.8, was reached for 2 hidden layers and 14 neurons per layer. Therefore, a multi-layer structure was defined with two hidden layers, one input layer, and one output layer (14 neurons per hidden layer). The input layer represented the selected features by the random-forest part, and the output layer corresponded to the free-electron concentration measured by the Hall measurement. Rectified Linear Unit (ReLU) was the activation function implemented in this neural network, and the Adam optimizer [42] was chosen since it is currently the most widely used gradient-descent algorithm.

The prediction performance of the fDNN model also depends on both hyper-parameters of the random forest and the neural network. The hyperparameters of the random forest include the number of trees in the forest and tree-related parameters such as the depth of the decision tree, the maximum number of features considered for splitting in a node, etc. The hyper-parameters of a neural network are framework-related parameters such as the number of hidden layers and the number of neurons per hidden layer and training-related parameters such as the learning rate and the batch size. The values of the above hyperparameters values were optimized by a combination of manual exploration and grid search, and the finalized values can be found in the Supplementary Materials Table S2.

In the research of thin-film development, one may not simply be satisfied with a predictive model for a specific system but may also be interested in finding the key descriptor for further process improvement. After fitting the fDNN model, a newly developed variable-ranking mechanism [29], which combines the variable-importance calculation embedded in random forests and the Connection Weights (CW) method [43] used in neural networks, was applied to calculate a score for each process parameter as the variable importance in the fDNN.

3. Results and Discussion

With the help of the variable-ranking mechanism, we first examined the influence of the initial input parameters (6 input parameters) on the doping level. The Ga precursor molar flow played a significant role in predicting, followed by the Ar push-gas flow, chamber pressure, and oxygen flow, as shown in Figure 2a. Surprisingly, the TEOS molar flow did not significantly influence the doping level according to the variable-ranking mechanism, which conflicts with our domain knowledge but can be explained by the multicollinearity of the TEOS molar flow values, as shown in Supplementary Materials Figure S1. The multicollinearity does not influence the prediction accuracy of the randomforest model but may deteriorate the interpretability of the variable-ranking result [44,45]. Therefore, the importance of the TEOS molar flow should still be highly evaluated for further model training. As reported in the literature, the Ga precursor is the dominant parameter in predicting the growth rate of the β -Ga₂O₃ film [46]. Following the observed similarity, the growth rate (nm/min) was introduced into the dataset, and a new parameter was found to be a decisive descriptor for the doping prediction, as shown in Figure 2b. This newly found parameter, the Si supplied per nm (mol/nm), can be defined by dividing the concentration of Si precursor (mol/min) with the thin-film growth rate (nm/min), which can be considered as the amount of Si dopant supplied while growing 1 nm of β -Ga₂O₃ film.



Figure 2. The parameter importance based on the trained fDNN model. The importance is ranked by the features containing the most information for the model to make a prediction of the free-carrier concentration in the films: (**a**) individual process parameters and (**b**) added with a created parameter: Si supplied per nm (mol/nm).

With this new parameter, a predictive model for (100) β -Ga₂O₃ thin film was developed by including the Si supplied per nm and the other process parameters (7 input parameters). A comparison between the predicted level and the measured doping level is presented in Figure 3. A robust linear relationship between the predicted values and measured values from the training and testing sets indicates that the fDNN model generalized very well to individual datasets, providing the training set/testing set R² of 0.99/0.86 and the RMSE of $1.4 \times 10^{16}/1 \times 10^{18}$. The prediction region of the developed model covered the range from 10^{17} to 10^{19} cm⁻³. The prediction deviation, especially at a heavily-doped regime (>1 × 10¹⁸ cm⁻³), mainly resulted from the noise of the measurement data and a relatively finite experimental resolution. By estimating the normalized root mean square error value (NRMSE), the prediction deviation was low enough for the practical estimation (NRMSE values are ≤0.1 for both training and testing set; NRMSE will be 0 in an ideal case), and it satisfied the limited size of the available dataset.



Figure 3. The plot of the predicted doping level versus the measured free-carrier concentration from (100) β -Ga₂O₃ thin films using the trained fDNN: (**a**) Training set and (**b**) Testing set.

Ten experiments on (010) oriented substrates were also performed in order to examine the model's validity for the substrate of different facet orientations (as shown in Table 2). A comparison between the predicted and measured doping levels is provided in Figure 4. The prediction error is in an acceptable range, which implies that the developed model was flexible enough to provide desirable prediction accuracy for both (100) and (010) substrates considering the system fluctuation of the MOVPE system. The possible prediction deviation of the doping level may be due to the lack of reliable in situ growth-rate-measurement techniques, as is widely utilized for the homoepitaxial growth of III-V group semiconductor materials [47], and the unknown incoming precursor molar flow on the substrate surface. It puts doubts on the precision of the collected growth parameters and the quality of data.

A model trained with a (100)-based dataset providing the generality to predict the doping level of (010)- β -Ga₂O₃-grown films suggests that the doping process in both substrate orientations may have no fundamental difference and share the same descriptor for the doping process. The R² of the predicted values for the (010) samples was 0.93 while the RMSE was around 4 × 10¹⁷, which were comparable with the results of the (100) samples. To further validate the observation of the trained model, two samples, (100) and (010), were grown in the same chamber with the SIMS measurement results shown in Figure 5a,b. It can be seen that the measured Si concentrations were both around 4 × 10¹⁷ cm⁻³, which matches the result of the model. Si peaks with a concentration close to mid-10¹⁸ cm⁻³ or higher are visible at the substrate and film interface in both samples. Similar Si peaks at the interface between the substrate and the film have already been reported in the literature [13,48,49]. Different reasons have been suggested, such as remaining SiO_x on the substrate surface [49] or Si impurities inside the chamber [48], but no commonly agreed con-

clusion has been confirmed yet. A detailed study on this specific issue has been performed but is beyond the scope of this work, and the results will be presented elsewhere.

Table 2. Experimental parameters for the model validation on (010) β -Ga₂O₃ thin films. The corresponding process parameters are listed below. The experimental results and detailed mechanism in Table 2 are referred to Refs. [12,46].

No. Test	Growth Temperature (°C)	TEGa Molar Flow (mol/min)	Chamber Pressure (mbar)	TEOS Molar Flow (mol/min)	Growth Rate (nm/min)
1	800	$1.5 imes10^{-5}$	20	$9.8 imes10^{-10}$	5.1
2	800	$1.5 imes 10^{-5}$	20	$9.8 imes10^{-11}$	4.8
3	800	$1.5 imes10^{-5}$	20	$3.9 imes10^{-10}$	4.8
4	800	$1.5 imes 10^{-5}$	20	$1.2 imes 10^{-9}$	4.6
5	800	$1.5 imes10^{-5}$	20	$1.6 imes10^{-9}$	4.6
6	850	$6.1 imes10^{-6}$	5	$1.5 imes10^{-10}$	1.2
7	850	$9.2 imes 10^{-6}$	5	$1.3 imes10^{-10}$	1.7
8	850	$9.2 imes 10^{-6}$	5	$5.3 imes10^{-10}$	1.4
9	825	$9.2 imes10^{-6}$	5	$7.6 imes10^{-10}$	1.8
10	825	$9.2 imes 10^{-6}$	5	$1.9 imes 10^{-9}$	2.0



Figure 4. Measured values and predicted values of free-carrier concentration from (010) β -Ga₂O₃ thin films with the process parameters listed in Table 2.

A doping engineering strategy was developed following the results of the model, with the Si supplied per nm (mol/nm) as the main variable. To provide an estimation for doping engineering, we defined a concentration contrast, $\frac{C_{i+1}}{C_i}$, between different doping levels (*i* + 1 and *i*) with Si supplied per nm, N_i , as a single parameter as the following form Equation (1):

$$\frac{C_{i+1}}{C_i} = \frac{N_{i+1}}{N_i} \tag{1}$$

Figure 6a,b show the free-carrier concentration obtained by the Hall Effect measurement and the Si concentration obtained by the SIMS measurement versus the Si supplied per nm (mol/nm) with the data points of both the in-house-grown (100) substrates (black squares, labeled as IKZ) and commercial (010) (red circles, labeled as IKZ-Tamura) substrates from our dataset and the literature (blue triangles, referred to Ref. [15]) fitted by Equation (1) [15]. These findings indicate that the Si-doping behavior for β -Ga₂O₃ is



substrate-orientation-independent, precursor-type-independent (SiH₄ is the Si precursor used in the work of Bhattacharyya et al. [15]), and equipment-independent.

Figure 5. The Si profiles by SIMS of (a) (100) and (b) (010) in the same-chamber experimental run. The experimental conditions are: the growth rate is maintained at 0.9 μ m/h with the TEOS molar flow kept at 1.84 \times 10⁻¹⁰ mol/min. The red dashed line indicates the interface between the substrate and the grown film.



Figure 6. Comparison of the results of (a) Hall Effect measurement and (b) SIMS measurement from both (100) and (010) MOVPE-grown β -Ga₂O₃ thin films with the Si supplied per nm (mol/nm) as the main variable.

Studies on the temperature-dependent states indicated a two-donor level, with a shallower donor energy level ($E_{D1} = \sim 10 - 35$ meV) and deeper second donor ($E_{D2} = \sim 80 - 100$ meV) energy levels [49–51]. Si is a common n-type dopant in β -Ga₂O₃ [52], which is predicted by the first principle of density functional theory (DFT) [53] to be a shallow donor with its preference site as a tetrahedral site Ga(I) under oxygen-rich condition due to a lower formation energy. As a result, Si generally incorporates more at (E_{D1}), leading to efficient activation at room temperature, and shows no significant probability to substitute the oxygen site. Therefore, the free-electron concentration is expected to be proportional to the density of Si atoms supplied during the film growth. Besides, Equation (1) also indicates a kinetically controlled doping process and that the thin-film growth rate is faster than the diffusion rate of Si adsorbed onto the surface; with a fixed TEOS molar flow but varied growth rate, the amount of supplied Si atoms on the substrate surface is constant, but the surrounding gallium-oxide matrix changes, therefore, the actually-incorporated Si amount and free-electron concentration (when all incorporated Si are electrically active) is limited by the thin-film growth rate instead of the supplied amount of Si precursor. It is

a well-known fact that the thin-film growth rate is an implicit function of a set of process parameters, such as precursor concentration, chamber pressure, and growth temperature. Following the discussion above, the previously reported observation of the influence of a single process parameter [12–15,37] on the doping level can be considered to influence the thin film growth rate instead of doping behavior. With the current model, we demonstrated that the doping behavior in MOVPE-grown β -Ga₂O₃ thin film can be concluded by a single descriptor from a macroscopic view, i.e., Si supplied per nm (mol/nm), when the point defect is the only compensating factor in the film. In some other works, hydrogen was revealed to be a potential shallow donor [39,54]. However, it was not stable in a temperature-elevated environment, according to our experience.

4. Conclusions

This work demonstrated that a hybrid deep-learning model can precisely predict the doping level in MOVPE-grown Si-doped β -Ga₂O₃ thin film in the range of 10¹⁷ to 10¹⁹ cm⁻³. With the help of the model, a key descriptor, i.e., the Si supplied per nm (mol/nm), was identified to show a linear relationship with the doping level of both (100) and (010) β -Ga₂O₃ thin films. Furthermore, an empirical relationship was extracted in order to estimate the doping level with a good generality for different substrate orientations, dopant precursor types, and deposition systems. This will save time in process development at both the lab and industry levels.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/ 10.3390/cryst12010008/s1, Figure S1: The correlation between the TEOS flow rate (mol/min) and the free carrier concentration measured by Hall effect measurement, Table S1: R² of fDNN with 1–3 hidden layer and 9–16 neurons from 10-fold cross-validation, Table S2: The hyperparameter of fDNN: random forest and neural network.

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