Implementation of an artificial neural network to predict properties of MOVPE-grown AlGaN layers

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Abstract. A feed-forward artificial neural network was employed in order to predict the growth rate and composition of Al\textsubscript{x}Ga\textsubscript{1-x}N metalorganic vapor deposition-grown layers depending on the flow of a precursor gas (trimethylaluminum, trimethylgallium, and ammonia). The results showed a good agreement with experimental data. The obtained dependencies seem to be reasonable in the context of well-known effects: the presence of parasitic reactions between trimethylaluminum and ammonia and GaN decomposition at low ammonia (high hydrogen) flow.

1. Introduction

Modern electronics and optoelectronics repeatedly require new materials and more advanced implementation of materials that are already in use. III-N is one of these material systems. Despite the widespread using of AlGaN-based transistors and light-emitting diodes, there are still a lot of difficulties and unclear points.

Metalorganic vapor phase epitaxy (MOVPE) is the main technology used for the fabrication of III-N heterostructures. In contrast to molecular-beam epitaxy (MBE), the epitaxial growth by MOVPE occurs by gas-phase and surface chemical reactions, not by physical deposition. Complex organic compounds are used as precursor gases, therefore a large number of chemical reactions take place during the growth process, especially when growing ternary alloys like AlGaN. Moreover, the design characteristics (such as the type of MOVPE reactor, the inlet shape, etc.) can have an impact on the properties of epitaxial layers. Thus, computer simulation of the MOVPE process is a challenging task that requires experimentation in order to validate the model.

In this paper, we present the results of implementing an artificial neural network (ANN) in order to predict the composition and growth rate based on the growth conditions.

2. Artificial Neural Network Structure

Interest in machine learning and artificial neural networks has increased significantly in recent decades. Nowadays, ANNs are applied in various spheres of life from finance, healthcare and medicine to mobile apps and the entertainment industry. As for science, neural networks can be considered as a good alternative to physically-based models in cases when modeling that accounts for all ongoing processes is unnecessary, impossible, or simple requires too much time and resources. For example, ANNs have reportedly been applied for modeling reactive ion etching [1], growth of Si\textsubscript{3}N\textsubscript{4}...
films by plasma-enhanced chemical vapor deposition [2], and quality of MBE InAlGaAs-based structures [3].

Basically, an ANN consists of nodes (neurons) forming several ‘layers’: the input layer (which receives input data and transfers them into the next layers), the output layer (which shows the ‘answer’) and one or more ‘hidden’ layers (where the calculations are performed). Every single neuron receives input data \((X_1, X_2...X_n)\) from other nodes and applies the ‘activation function’ \(f\) to the weighted sum of the inputs (figure 1). Additionally, there is one more input 1 with a weight \(b\) called ‘bias’. Usually, the information moves only in one direction, forward, so this type of ANN is called a Feedforward Neural Network (figure 2).

The ANN’s learning process, called the back-error propagation algorithm [4], is as follows. The initial weights are set randomly, and the output is calculated. Then, the mean squared error between calculated and measured data is estimated. This error is subsequently minimized by tuning the weights using special algorithms.

3. Experimental design
AlGaN samples were grown by metal-organic vapour-phase epitaxy on (0001) sapphire substrates in a 6×2-inch planetary reactor of an AIXTRON AIX2000HT system. Trimethylgallium (TMGa), trimethylaluminum (TMAI) and ammonia (NH₃) were used as precursors; hydrogen was used as the carrier gas. The total gas flow and the reactor pressure were maintained at 28 standard liters per minute (SLM) and 100 mbar, respectively. NH₃, TMGa, TMAI and H₂ flows were chosen as the input factors. The growth rate and composition of the growing layers were measured by in-situ optical reflectance monitoring and chosen as output data. Further details on growth conditions can be found in Refs. [5] and [6].

The ANN was constructed using an MLPRegressor in the scikit-learn framework for the Python programming language [7]. It consisted of an input and output layer with a linear activation function and two hidden layers with a sigmoid activation function. The dataset was split into two subsets (training and validation) to avoid overlearning. The limited-memory Broyden-Fletcher-Goldfarb-Shanno (‘lbfgs’) algorithm was used for weight optimization since it performs better than the widely-used Stochastic Gradient Descent algorithm when the dataset is relatively small.

4. Results and discussion
It is known that the AlN growth rate decreases with increasing ammonia flow due to parasitic reactions between TMAI and NH₃. On the other hand, GaN tends to decompose in a hydrogen atmosphere, and increasing the proportion NH₃:H₂ suppresses its decomposition. Therefore, the GaN growth rate increases with increasing NH₃ flow. However, when the AlGaN growth process occurs (i.e. TMGa, TMAI, NH₃ and H₂ are present in the reactor at the same time), these effects begin to interplay and affect each other. The dependencies become very complex, and taking every process into...
account in physically-based simulations becomes resource-intensive. The detailed experimental and theoretical investigations of these effects can be found in [5] and [8].

Figure 3. Growth rate and composition predicted by the ANN versus experimental data. Squares are the data from the training dataset; open circles are the data outside the training dataset.

Figures 3 and 4 show the comparison between experimental data and ANN predictions. After making allowances for the measurement accuracy, one can say that they are in very good agreement. Figure 5 represents the predicted dependencies of the layer composition on the NH₃, TMAI and TMGa flows. When the NH₃ flow is relatively small, it is almost impossible to get an AlₓGa₁₋ₓN layer with \( x < 0.4-0.5 \) due to the decomposition of GaN in an atmosphere with a high hydrogen content (figure 5a). Under a high ammonia flow, in contrast, an AlGaN layer with a high aluminum content can be fabricated only when the TMGa flow is moderate (or even low), wherein the amount of TMAI flow plays almost no role. The obtained dependencies seem reasonable.

Figure 4. Dependency of the AlGaN growth rate on the precursor flows. Open circles are the data outside the training dataset. TMAI and TMGa flows are in µmol/min.
Figure 5. Predicted composition versus the flows of precursor gases.

Figure 6. Predicted growth rate versus the flows of precursor gases. The inset shows the growth rate profile along the red line (TMGa flow is 200 µmol/min).

The growth rate dependency on precursor flows is shown in figure 6. When the flows of TMAI and ammonia are low (i.e. the hydrogen concentration is high), the growth rate is about zero due to the GaN decomposition. An increase in the TMAI and TMGa flows causes the growth rate to increase at low and moderate NH₃ flows. However, when the ammonia flow is high, a situation might arise when an increased TMAI flow results in a decreased growth rate (figure 6c). This could be related to a strong increase in gas-phase parasitic reactions between NH₃ and TMAI. A high concentration of
TMAl an NH₃ in the gas phase leads to depletion of the effective volume of precursors and, thus, to a decrease in the growth rate in spite of the increase in the TMAl flow. This effect was observed in other experiments and is in a good qualitative agreement with predictions.

5. Conclusions
An artificial neural network model was used to predict the composition and growth rate of an AlGaN layer in a MOVPE reactor depending on precursor flows. The predicted values were found to be in good agreement with the experimental data. The obtained dependencies seem to be reasonable in the context of the presence of parasitic reactions between TMAl and ammonia and GaN decomposition at a low ammonia (high hydrogen) flow. Using the ANN model can help to find the optimal growth conditions. The implementation of additional input parameters (e.g. the substrate temperature, pressure, etc.) or output parameters (e.g. defect density or RMS roughness of the surface) would help find even better conditions for faster growth of high-quality AlGaN layers of the desired composition.

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