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Using optical reflectance to measure GaN nucleation layer decomposition kinetics

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Abstract

GaN nucleation layers (NLs) that are used in the two-step GaN growth process on sapphire partially decompose during the ramp from low to high temperature, T. In this paper, we show how the extent of this decomposition can be measured during each growth run using optical reflectance. Using reflectance, the NL decomposition rates were observed to increase with increasing P and T, and decreasing NH_3 flow. Also, the NL decomposition rate did not change as the NL growth T was varied, but did increase as the NL thickness increased. The NL decomposition has an activation energy, E_A, of 2.7 ± 0.2 eV for all annealing conditions studied, while the pre-exponential factor, A_0, depends strongly on the annealing conditions and initial NL thickness. Using the measured NL decomposition kinetics, an analytical model is used to fit the reflectance waveforms which also includes the T dependence of indices of refraction. The model was applied to several sets of reflectance waveforms where the annealing conditions were intentionally varied and the values of A_0 measured and compared.

In addition to directly fitting the reflectance waveforms, we introduce another way to quantify decomposition rates measured under constant or varying T. This approach requires a constant E_A for the reaction kinetics throughout the entire T range of interest, which is the case for the GaN NL decomposition. For this approach, time is scaled by an integrated form of the Arrhenius exponential (i.e. $\exp(-E_A/k_B T)$). We have called this transformation of the time scale the "kinetic advancement," because it allows kinetic processes at constant or varying T to be compared directly. If the reflectance waveform or NL thickness is plotted vs. the kinetic advancement, a straight line results with the slope equal to A_0. The usefulness of both reflectance waveform fitting procedures is discussed, specifically how the reflectance waveform can be compared and quantified during every growth run in order to verify NL evolution repeatability.

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1. Introduction

Prior to the use of nucleation layers (NLs), GaN films grown directly on sapphire resulted in material with high defect densities. The use of NLs as first demonstrated by Amano and coworkers using AlN in 1986 [1] and later in 1991 by Nakamura [2] and Wickenden et al. [3] using GaN, improved both the optical and electrical properties compared to films grown directly on sapphire [1]. Following this pioneering work, many groups have developed GaN growth capability on sapphire using AlN and GaN NLs and today these NLs are used routinely for the fabrication of UV, blue and green LEDs as well as other nitride-based optical and electronic devices.

Typically, GaN NLs are deposited between 500 and 600 °C, after which the NL is ramped to 950–1100 °C for continued high temperature, T, growth. During the temperature ramp changes occur in both the morphology and crystalline quality of the NL. These changes include loss of material [4], reincorporation of gas phase atoms into the developing nuclei [5], improvements in the NL crystalline quality [6], and an increase in roughness [7]. By varying the annealing schedule [8,9] and environment as shown here, the NL decomposition kinetics and subsequent NL evolution can be significantly altered.

How the NL evolves during the T ramp before high T growth is still somewhat unclear. It is known that GaN nuclei are generated [5] and it is on these nuclei that the subsequent high T GaN growth occurs. Obviously, how the nuclei form and how the high T growth proceeds influence the microstructure and subsequent dislocation density [10,11]. Ideally, methods to measure and control the extent of NL decomposition and nuclei formation during each growth run are strongly desired.

In this paper, we expand on our previous measurements of NL decomposition using optical reflectance. To calibrate the reflectance signal to the NL thickness, a model was developed to fit thick NLs. This model is based on the virtual interface method [12] and includes a term that accounts for surface roughening. Next, decomposition results are presented as a function of the T, NH_3 flow, pressure, NL growth T, and NL thickness. After this, we show how the reflectance waveforms can be modeled using the measured NL decomposition kinetics. The model involves iterative fitting by changing only the pre-exponential factor, A_0. Another method is then presented which rescales time so that the reflectance waveforms can be plotted to yield A_0 directly. Examples using both fitting procedures are then compared. Finally, we discuss how the results presented in this paper compare to previous studies, and demonstrate how the NL decomposition kinetics can be measured during each growth run using optical reflectance.

2. Theoretical development

In this section modifications are made to the virtual interface method [12] so that reflectance waveforms can be simulated for the group III nitrides. Because GaN typically roughens before coalescing, we assume here for simplicity that the reflectance attenuation occurs due to simple geometric roughness. Lastly, we describe a method that rescales time so that reflectance waveforms with constant or varying T can be compared directly.

2.1. Fits to the optical reflectance waveforms

In 1995, Breiland and Killeen developed a virtual interface method to model normal incidence optical reflectance [12]. With this method the growth rate, G, and the high temperature optical constants, n and k, could be extracted during growth. With this model, simplifying assumptions were made on the range of refraction indices ($n>4$) and on the adsorbing nature ($k>0$) of the films and substrates. Using these two simplifying assumptions, a damped cosine expression was derived that was applicable for most semiconductor growth, as shown in Eq. (8) of Ref. [12].

However, for GaN and sapphire the indices of refraction, n, are 2.3–2.4 [13] and 1.77 [14], respectively, i.e. $n<4$. Also over the wavelengths 400–1000 nm commonly used for reflectance, $k=0$ for both GaN and sapphire, i.e. they are