Monitoring the formation of nanowires by line-of-sight quadrupole mass spectrometry: a comprehensive description of the temporal evolution of GaN nanowire ensembles

Sergio Fernández-Garrido,* Johannes K. Zettler, Lutz Geelhaar, and Oliver Brandt

Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5–7, 10117 Berlin, Germany

E-mail: garrido@pdi-berlin.de

Supporting Information

Additional information on the line-of-sight quadrupole mass spectrometer

In order to measure the desorbing Ga flux during growth, a Pfeiffer-Vacuum PrismaPlus quadrupole mass spectrometer equipped with a continuous secondary electron multiplier detector was installed in one of the cell ports of the molecular beam epitaxy system. The angle of the cell port with respect to the substrate normal is 44°. Figure S1 shows a sketch of the line-of-sight quadrupole mass spectrometer (QMS) setup. As shown in the figure, we introduce an aperture between the QMS ionizer and the substrate to restrict the line-of-sight acceptance angle. The distance between the QMS ionizer and the center of the wafer is 596 mm. An aperture with a diameter of 8 mm placed at 296 mm from the ionizer accepts only those Ga atoms desorbed from the inner 1.5 inch of the wafer. As described in Ref. 25, the quadrupole response to the Ga^{69} signal was calibrated in GaN-equivalent growth rate units by measuring the Ga^{69} signal when a $Al_2O_3(0001)$ substrate is exposed to different Ga fluxes at a temperature where all impinging Ga atoms are desorbed. As shown in Fig. S2, the Ga^{69} signal increases linearly with the impinging Ga flux. The QMS response was thus derived from a linear fit to the experimental data.

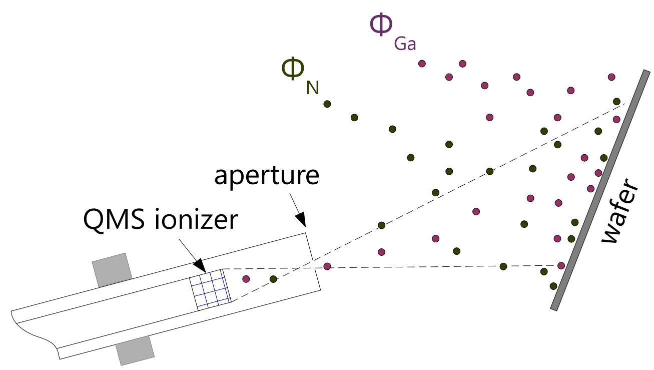

Figure S1: Schematic diagram of the QMS system.

Summary of the samples used in this work and of the constants derived from QMS transients

Table S1 summarizes the growth conditions used for all the samples presented in this work and table S2 the time constants, t_1, τ_1, t_2 and τ_2 derived from fitting eq.1 to the QMS transients.

*To whom correspondence should be addressed

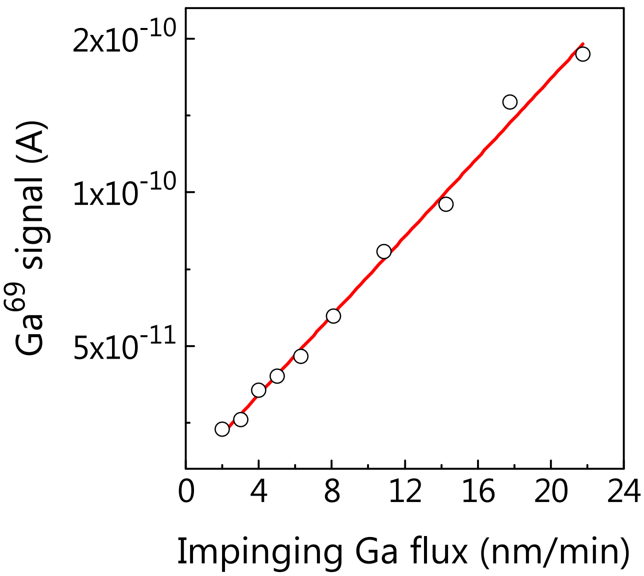

Figure S2: Ga^{69} signal measured by QMS as a function of the impinging Ga flux on a $Al_2O_3(0001)$ substrate at 910°C. The solid red line is a linear fit to the experimental data.

Temperature dependence of τ_2

As shown in Fig. S3(a), the substrate temperature has a strong impact on the parameter τ_2, which determines the time variation in the contribution of collective effects to the total deposition rate. As can bee seen in the figure, τ_2 increases from 12 to 34 min when the temperature is increased from 775 to 835°C. However, in contrast to the delay time for the onset of collective effects t_2, the temperature dependence of τ_2 follows an Arrhenius law only with a large error margin. The activation energy derived from the fit is (2.0 ± 0.4) eV.

Impact of the impinging Ga flux on t_2 and τ_2

Figure S3(b) presents the variation with the impinging Ga flux of the delay time for the onset of collective effects t_2 and the time constant τ_2. For both parameters we observe a clear trend, namely, the higher the Ga flux the shorter the times. As in the case of the parameters related to the nucleation stage, t_1 and τ_1, the dependencies of t_2 and τ_2 on the impinging Ga flux can be described by sim-

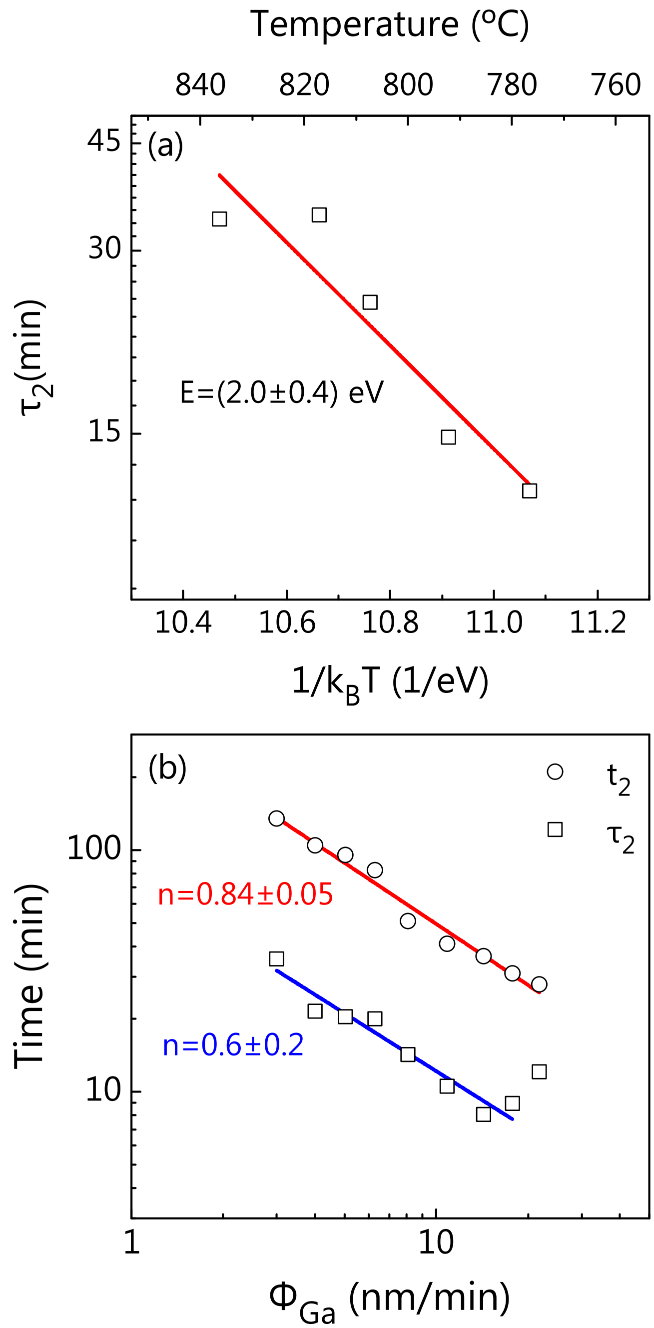

Figure S3: (a) Temperature dependence of the time constant τ_2 for a series of samples grown with $\Phi_{Ga} = (4.9 \pm 0.6)$ nm/min and $\Phi_N = (10.5 \pm 0.5)$ nm/min. The solid line is a fit of the data by an Arrhenius law. (b) Variation with Φ_{Ga} of the delay time for the onset of collective effects t_2 and the time constant τ_2 for a series of samples grown at 805°C with $\Phi_N = (10.5 \pm 0.5)$ nm/min. The solid lines are fits of the data by eq. S1.

ple power laws defined as:

$$t_2, \tau_2 \propto \frac{1}{\Phi_{\mathrm{Ga}}^{n}}. \tag{S1}$$

The values of the exponents n derived from the fits shown in Fig. S3(b) for t_2 and τ_2 are 0.84 ± 0.05 and 0.6 ± 0.2, respectively. These results evidence that the time required to form a homogeneous nanowire ensemble depends not only on the substrate temperature but also on the impinging Ga flux.

Table S1: Impinging fluxes Φ_{Ga} and Φ_{N} in nm/min, substrate temperatures T in °C, and growth times t in min for all the samples (#) presented in this work.

#	Φ_{Ga}	Φ_{N}	T	t
1	4.9±0.6	10.5±0.5	805	15
2	4.9±0.6	10.5±0.5	805	30
3	4.9±0.6	10.5±0.5	805	45
4	4.9±0.6	10.5±0.5	805	60
5	4.9±0.6	10.5±0.5	805	85
6	4.9±0.6	10.5±0.5	805	110
7	4.9±0.6	10.5±0.5	805	130
8	4.9±0.6	10.5±0.5	805	180
9	4.9±0.6	10.5±0.5	775	180
10	4.9±0.6	10.5±0.5	790	180
11	4.9±0.6	10.5±0.5	815	180
12	4.9±0.6	10.5±0.5	835	450
13	3±0.2	10.5±0.5	805	210
14	4±0.5	10.5±0.5	805	180
15	6±1	10.5±0.5	805	180
16	8±1	10.5±0.5	805	180
17	11±2	10.5±0.5	805	180
18	14±2	10.5±0.5	805	180
19	18±2	10.5±0.5	805	180
20	22±3	10.5±0.5	805	180
21	4.9±0.6	4.7±0.5	815	200
22	4.9±0.6	10.5±0.5	815	200

Table S2: Time constants t_1, τ_1, t_2, and τ_2 in min derived from fitting eq.1 to the corresponding QMS transients.

#	t_1	τ_1	t_2	τ_2
8	41	4.6	90	23
9	6	1.8	29	12
10	16	3	48	15
11	80	7.4	132	34
12	252	10.9	325	34
13	82	10	135	36
14	53	7	105	22
15	37	5.8	82	20
16	20	3.5	51	14
17	14	2.6	41	11
18	9	2.1	37	8
19	6	1.4	31	9
20	4	1.8	28	12