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Kinetics and statistics of nanowire growth

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The formation of nanowires (NWs) of semiconductors and of their heterostructures offers several choice examples of crystal growth in a confined medium of nanometric size. I shall discuss different studies carried out recently at LPN in the domain of III-V nanowires, concentrating on the intimate coupling between the modeling of epitaxial growth and its experimental study. This will give me the opportunity to recall how nanowires grow, without hiding that the understanding of the growth mechanisms, and sometimes even their very identification, are far from being stabilized yet.

First recall that NWsfrequently grow in the vapor-liquid-solid (VLS) mode, from a liquid (L) catalyst droplet whose volume is of the order of a zeptoliter. The droplet is fed by the vapour (V) fluxes set up in the growth chamber and growth produces the solid (S) NW.

1. Self-catalyzed growth

Modeling VLS growth of III-V NWs mediated by a foreign catalyst (such as Au) is made tricky by the complex composition of the droplet and the multiplicity of material pathways involved. Modeling self-catalyzed growth, where the catalyst consists of the group III element of the solid NW, is potentially simpler. In particular, the thermodynamics of the (III,V) liquid that constitutes the droplet are known. And for GaAs at least, a striking feature of this method is that the NW growth rate is governed by the group V (As) flux supplied to the sample and varies linearly with it. At the same time, As diffusion is negligible. We have thus developed an “As-only” fully quantitative model of the self-catalyzedmolecular beam epitaxy (MBE) growth of GaAsNWs, that depends on only a few a priori unknown physical parameters [1]. The model relies on As material balance between the three VLS phases and includes nucleation, evaporation and capture

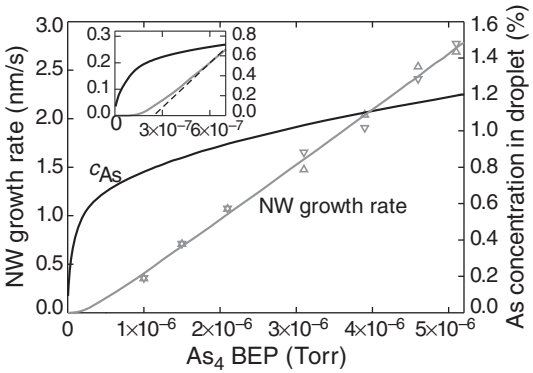

Figure 1. Comparison of growth rates measured on two NWs as a function of As_4 beam equivalent pressure (BEP) (triangles) with modeled growth rate (red line), and calculated atomic composition cAs of the droplet (blue line). The inset details the calculated variations of the growth rate and As concentration at low incident fluxes (same units as main panel); in this range, the growth rate deviates markedly from the linear approximation valid at higher pressure (dashed line).

of As re-emitted by the NW environment. It reproduces quantitatively all salient features of our experimental study [2], in particular the variations of NW growth rate with As flux (Fig. 1), temperature and NW radius.

From these optimized fits (Fig. 1), we extract, for the first time in the field of NW growth, a complete set of model parameters, in particular the nucleus edge energy. We also determine quantities so far inaccessible to experiment, such as As concentration in the Ga droplet (about 1%; Fig. 1), liquid supersaturation and nucleation barrier, for individual NWs. The model can then be used to predict the growth rate (and all quantities of interest) for arbitrary GaAs NWs under arbitrary growth conditions, including conditions not yet explored experimentally (Fig. 1, inset). Although largely ignoring group III elements, our model thus seems to capture most of the physics of self-catalyzed growth.

2. Sub-Poissonian statistics of nucleation from a nanophase

In VLS growth, the depletion of the nanodroplet that occurs when a new solid nanowire monolayer is formed induces very original nucleation statistics and confers a self-regulated character to growth.

Namely, in 2010, we demonstrated experimentally the existence of sub-Poissonian nucleation statistics in VLS NW growth and modelled them numerically [3]. The striking feature is that the distribution of the numbers of nucleation events occurring in fixed time intervals is much narrower than the Poissonian distribution

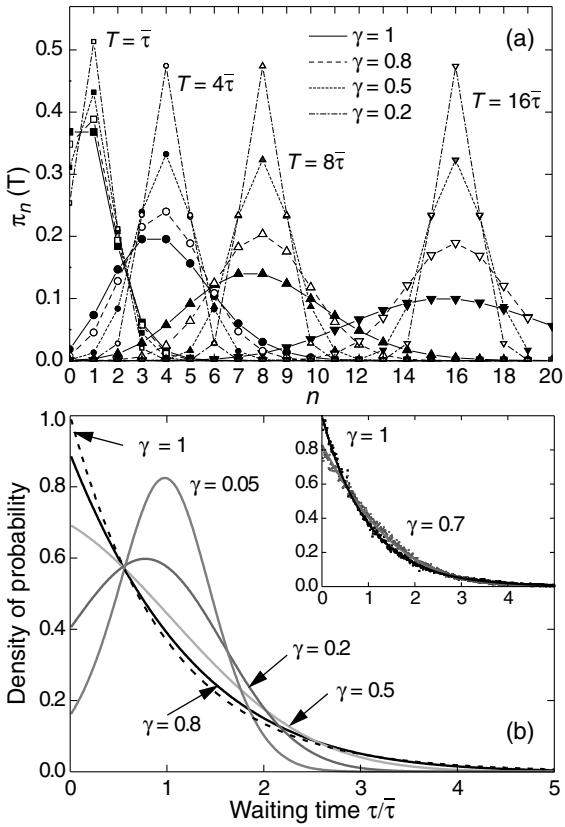

Figure 2. (a) Distribution of the probabilities of having nnucleations in a time T equal to 1 (squares), 4 (circles), 8 (up triangles) and 16 (down triangles) average waiting times $\bar{\tau}$ for the Poisson process ($\gamma = 1$; large full symbols, full lines) and various sub-Poissonian processes ($\gamma < 1$) The lines are only guides to the eye. (b) Probability distribution of the waiting time τ (normalized to $\bar{\tau}$). Inset: comparison of analytical calculations (lines) with numerical simulations (dots).

that would be observed for non-correlated events. We also argued at that time that the observed temporal anti-correlation (or self-regulation) of the nucleation events results from the depletion of the droplet due to the rapid formation of a NW monolayer (ML) triggered by nucleation. We have now developed a fully analytical calculation of these statistics [4]. This allows us to discuss their various manifestations and to make quantitative comparisons with experiments.

The sequence of nucleation events is modeled as a stochastic Markov process. The deviation from Poisson statistics is quantified by a single parameter γ, the ratio of the nucleation probabilities immediately before and after nucleation. The basis of our calculation is a self-consistent determination of the densities of probability of the nucleation probability, both when nucleation occurs and at an arbitrary instant. From these, we compute various quantities amenable to comparison with experiments or simulations.

Namely, we first derive the probability for having a given number of nucleations in a given time interval. As noted before, the distribution of these probabilities shows a marked narrowing with respect to Poisson statistics. We then calculate explicitly the standard deviation of this distribution, which quickly saturates as the length of the time interval increases. These results are compared with post-growth measurements. We also compute the distribution of the waiting times between nucleations, which could only be measured during growth.

We discuss how the computed quantities vary with parameter γ. We derive the actual values of γ for Au-catalyzed and self-catalyzed growth. A remarkable conclusion is that a marked narrowing of the distribution of the numbers of nucleations occurring in fixed time intervals [Fig. 2(a)] is fully compatible with a very broad distribution of waiting times [Fig. 2(b)]. This means that even strongly sub-Poissonian statistics do not at all imply that nucleation occurs in a quasi-periodic fashion. We discuss the implications of these findings for the realization of precisely controlled heterostructures in NWs.

I will finally consider the implications of these peculiar statistics on the formation of precisely controlled quantum structures in NWs.

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Rydberg excitons in cuprous oxide

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Cuprous oxide is the material in which excitons were discovered first by Evgenii Gross and Nury Karryjew in 1952. The energy levels of excitons typically can be well described by the hydrogen series. In the talk I will discuss the recent