Semiconductor Nanostructures Self-Organized by the Turing Instability

Jiro Temmyo and Toshiaki Tamamura

NTT Opto-electronics Labs, 3-1 Morinosato-Wakamiya, Atsugi, Kanagawa 243-01, Japan Phone: +81 462 40 2855, Fax: +81 462 40 4305, Electronic mail: temmyo@aecl.ntt.co.jp

1. Introduction

We propose a new interpretation of strained InGaAs quantum disks [1-2] which appear spontaneously during the epitaxial growth on a GaAs(311)B surface. In this paper, we describe the surface topography domain in a strained InGaAs/AlGaAs heterostructure system on the GaAs (311)B face, suggesting that the surface rearrangement process is self-organization induced by the Turing instability [3].

2. Turing instability

Turing structures [3] are stationary periodical concentration patterns resulting from a diffusive instability originating from the sole coupling of reaction and diffusion processes in nonlinear dynamical systems. They correspond to stable stationary solutions of a set of reaction-diffusion equations (nonlinear partial differential equations). Here, we focus on the resemblance between our regularly-dotted InGaAs morphology on the GaAs (311)B face and the spotted skin of the cheetah (Figures 1(a) and (b)). Surprisingly, a singleTuring pattern-formation mechanism may possibly underlie the wild variety of animal coat markings found in nature although it is not clear as to precisely what happens during embryonic development to cause the pattern. If this analogy were correct, the so-called "reactiondiffusion mechanism" predicted by Turing may also dominate the epitaxial growth process on the (311)B surface.

3. Experimental

The samples were grown in a vertical low-pressure metalorganic-vapor-phase-epitaxy (MOVPE) reactor using rf heating. The crystal faces of the GaAs substrates used here were (311)B and (100). The typical epitaxial structure consists of a 10-nm-thick GaAs buffer layer, a 30-nm-thick AlGaAs layer, an InGaAs initiating layer, a 100-nm-thick lower AlGaAs barrier layer, a thin InGaAs layer, a 50-nm-thick upper AlGaAs barrier layer, and a thin InGaAs cap layer. After the thin InGaAs inner layer and the InGaAs capping layer were deposited, we interrupted the growth under the arsine atmosphere for 3 minutes. The surface images were analyzed with a highresolution scanning-electron-microscope (SEM).

4. Results and Discussion

Typical SEM topographical images of as-grown, 10-nm thick In0.2Ga0.8As/AlGaAs on GaAs (311)B faces are shown in Fig. 2. Figures 2(a)-(c) show results obtained at various substrate temperatures during the growth interruption. When we maintained the temperature at 800°C, we found well-defined, well-ordered arrays of nanocrystals in a triangular lattice. As discussed in detail in Refs. 1 and 2, these nanocrystals contain the built-in InGaAs quantum disks. When the temperature was maintained at 750°C, however, we found a complex, modulated surface pattern, including a honeycomb lattice and a reciprocal triangle lattice. When the substrate temperature was maintained at 700°C, we find only a flat morphology. For the (100) face, not shown here, a flat morphology was found for each of these growth-interruption temperatures. Figures 2(d)-(f) also show a series of surface patterns of In0.2Ga0.8As on the GaAs (311)B faces. Note that the formation of nanocrystals is favored on underlying AlGaAs layers containing a strained InGaAs initiating layer and note the suppression of the process at the lower substrate temperatures, and in the presence of thinner InGaAs layers. Here, we call these morphologies in Figs. 2(d)-(f) a pattern of periodically zigzag-striped nanocrystals.

Completely-isolated nanocrystals of different sizes are also obtained. Basically, the disk size of the triangle-lattice nanocrystals can be roughly controlled in the range of 150 nm to 30 nm by changing the indium composition. The estimated wavelength, (i. e., the period of the neighboring nanocrystals) is between 450 and 130 nm. Another series of experimentally modulated topographies are obtained with higher indium compositions. We can find the spontaneous emergence of pronounced zigzag-striped patterns that include some nanocrystals. Note that for the indium composition of 30%, the growth condition is so critical that the triangle-lattice or the zigzag-striped pattern is obtained for the run-to-run growth. The honeycomb lattice was almost never obtained when the indium content was high. Figure 3 roughly summarizes the features of the surface morphologies. This surface topography domain might correspond to the Turing space (a steady spatially inhomogeneous state) as discussed later.

The mechanism of this rearrangement seems to be quite different from that of the conventional S-K (Stranski-Krastanow) growth mode of the simple self-assembled islanding phenomenon [4].



Fig. 1. Comparison between a surface image of as-grown InGaAs on a GaAs(311)B and a spotted pattern of the cheetah (taken by M. Iwago).

It is worthwhile noting here that if appropriate kinetics, such as auto-catalysis and substrate inhibition mechanisms, are involved, the uniform state can become unstable when a control parameter crosses a critical value, and various structures can spontaneously develop according to a system of reactiondiffusion equations (Turing instability) [3]. The existence of the large difference in the diffusion constant of at least two species is needed for the Turing instability. In contrast to the flat homogeneous pattern of InGaAs on the GaAs (100) face, these results may be due to unique features of the GaAs (311)B surface. There exist two kinds of sites on the (311)B surface: the double dangling site seen in the [100] direction, and the single dangling bond site seen in the [111]B direction. Their densities are exactly the same. In addition, there are no atomic steps along the [011]. Because of the arsenic-stabilized surface due to the single dangling-bond of arsenic, it is very difficult to grow epitaxial layers such as GaAs and AlGaAs films on the GaAs (111)B substrate [5]. Note that the formation process on the (311)B face is somewhat related to the difference of the adsorption rates on the (100) and (111)B faces. Accordingly, our physical rearrangement process seems to involve antagonistic activatory and inhibitory kinetic processes induced by the (100) and (111)B faces. The intermixing process seems to correspond to the reaction process in the chemistry. We can translate the chemical term "reaction-diffusion system of the molecules" to the crystal growth term "intermixing-diffusion system of the adatoms." In particular, a species controlling the inhibitory process must diffuse faster than a species controlling the activatory process. Here we are assuming that the intermediate products of our system are InGaAs disks formed and InGaAs intermixed by AlGaAs. However, the actual diffusion constants for the two species are not known at the present.

When we take the feature of the atomic arrangement symmetry of the (311)B face into account, the pattern selforganized on the surface can be considered to be principally a triangular lattice, which includes tetragonal and hexagonal lattices with modified penta-hepta lattices. This pattern seems to be a result of the competition of the meta-stable rectangular and stable hexagonal lattices.

5. Conclusion

Unique surface morphologies on the (311)B face suggest a novel fourth growth mode due to Turing-type self-organization on the high-Miller-index faces. The experimental evidence of the possible Turing instability in the strained epitaxial growth creates a new perspective not only for nanostructure fabrication technology, but also for fields such as nonequilibrium physics and nonlinear mathematics.

Acknowledgments

The authors wish to thank R. Nötzel for his SEM analysis in the initial stage.

References

1. R. Nötzel, J. Temmyo and T. Tamamura: *Nature* **369** (1994) 131. 2. J. Temmyo, R. Nötzel, H. Kamada, T. Furuta and T. Tamamura: *Proc. 22nd Int. Conf. Phys. of Semicond., Vancouver*, 1994 (1994) p.1851.

3. A. M. Turing: Phil. Trans. R. Soc. B237 (1952) 37.

4. L. Goldstein, F. Glas, J. Y. Marzin, M. N. Charasse and G. Le Roux: Appl. Phys. Lett. 47 (1985) 1099.

5. K. Kato, Y. Hasumi, A. Kozen and J. Temmyo: J. Appl. Phys. 65 (1989) 1947.







Fig.2. SEM surface images of as-grown thin In0.2Ga0.8As sequentially grown on, typically, Al0.5Ga0.5As for the substrate temperature of (a) 800°C, (b) 750°C, (c) 700°C, (d) 800°C without an initiating layer, (e) 800°C with a In0.2Ga0.8As thickness of 5-nm on Al0.2Ga0.8As, and (f) 750°C with a In0.2Ga0.8As thickness of 5-nm. A SEM cross-sectional view included for (a), indicates the existence of a very small confined nanostructure of an InGaAs quantum disk.



Fig. 3. Summary of surface morphologies of as-grown InGaAs as functions of indium composition and inverse temperature. This surface topography domain might correspond to the Turing space (a steady spatially inhomogeneous state).