Report on the thesis presented by

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Entitled

Theoretical studies of rolled-up and wrinkled nanomembranes

The thesis presented by Mr. Cendula deals with the theoretical prediction of the three-dimensional shape of thin bi-layer semiconductor films. The films are initially bonded to the substrate and contain internal stresses due to a difference in layer strains. When the films are released from the substrate by removal of a sacrificial layer (mostly by etching), the films relax and attain their three-dimensional shape by wrinkling or rolling. In the thesis a theoretical/numerical approach is followed to predict the shapes formed in experiments and to elucidate the competition between rolling and wrinkling and the preferred dependence on internal strain, outer film shape and material properties. In addition, the effect of film shape and the associated internal strains on the electronic and optical properties are studied.

The thesis contains an abstract and an introduction in which the scope of the thesis is discussed and placed in the wider perspective of microsystem fabrication at the nano-scale as currently required for NEMS and MEMS applications in micro-electronic and photonic systems. In chapter 1 an overview is given of small-strain plate theory as well-known form the standard text books. In addition, the large strain extension is described according to Fopple-von Karman plate theory. The strain energies are presented in terms of integral formulations and the associated Euler equilibrium equations are derived from the integral equations by minimization. These equations form the basis for the analysis in the subsequent chapters. Chapters 2, 3 and 4 contain the scientific content of the paper based on three journal; publications, of which two have been published and one is submitted. The thesis is accompanied by an extensive bibliography (160 references), which is up-to-date and places the current work in a proper scientific perspective, and a list of symbols and notations.
(Nomenclature). No general discussion or outlook is presented at the end of the thesis. Instead, each chapter contains a discussion and outlook on the specific sub-problems studied.

Chapter 2 starts with a discussion on the competition between rolling and wrinkling as observed in several experimental systems. In chapter 2.1, the total strain energy is calculated in the case a freely hanging bilayer film is rolled-up in a tube and subsequently the energy is minimized, giving the equilibrium strain and equilibrium radius of the rolled-up tube. In case the film strains are much larger, the film-stresses will not relax by rolling but by wrinkling. For this situation Mr. Cendula calculates the total wrinkle energy in terms of the stretching and bending energy. Then, the energy is minimized giving the equilibrium amplitude and wavelength that comes about due to the competition of bending and stretching contributions. Subsequently, in chapter 2.3, the results of the two analyses are compared, resulting in a deformation mechanism map, depicting the regions in which wrinkling or rolling is dominant for a given average strain. In the map also the tube radius and wrinkle wavelength are depicted by means of contour plots for the respective regions of dominance. In chapter 2.4 Mr. Cendula zooms in towards the theoretical prediction of the number of tube rotations, before the rolling process has been observed to stop experimentally. For this, it is assumed that a competition between wrinkling and rolling is responsible for stopping as illustrated in fig. 2.7. The results are presented in terms of similar mechanisms map for rolling showing the max. number of rotations for a given average strain and strain gradient (fig. 2.8). The chapter is closed with a short conclusion section and outlook.

In the modeling the films are assumed to be elastically isotropic, while in reality the deposited films are crystalline and therefore anisotropic. In addition, the presence of the substrate in the theoretical predictions is ignored, while it is well-known that they can impose strong cohesive forces to the films, affecting the film morphologies. How are these two aspects expected to affect the results?

In chapter 3 the focus is on the rolling of finite-sized films etched from all sides simultaneously. In that case rolling occurs by default along the long edge of the rectangular film. To overcome this the films are deposited on wrinkled sacrificial layers so that the bending stiffness is anisotropic. Energy-minimization is used to compare the strain energies for the cases where the film rolls into a tube along the long edge (i.e. perpendicular to the initial wrinkles) and rolls along the short wedge (i.e. parallel to the initial wrinkles). The results are summarized in a phase diagram in which the dominant rolling modes are identified as a function of strain gradient in the film and the amplitude of the initial wrinkles. Since additional energy is spent in overcoming the initially stored strain energy of the wrinkles, a regime can be identified where rolling occurs along the short edge (as opposed to the default long-edge rolling direction for initially flat films).

A small section is added on the rolling of graphene sheets. The physical origin of the strain gradient $\Delta \varepsilon$ in a carbon sheet of one atom thick remains unclear. How can the rolling be clarified in case of single-atom films?
Finally, in section 3.2 the theoretical results are compared with non-linear geometry finite element simulations (COMSOL), showing excellent agreement. The last part of chapter 3 contains a proof-of-principle experiment to verify the theoretical predictions. Wrinkled photoresist was deposited with a metallic layer to introduce a strain gradient. The experiments clearly show that rolled-up tubes can be fabricated by rolling along the short edge.

The thesis is closed by a theoretical analysis of the effect of film shape and the associated internal strains on the electronic and optical properties. Chapter 4.1 contains a review of the band structure of direct-gap semiconductors, focusing on the difference between compressively strained, unstrained and tensile strained direct band-gap semiconductors. As the deformation of the lattice changes the atomic spacing a change in band structure is induced. Also optical transitions are triggered when electrons are moved from valence to conduction bands, absorbing light, and vice versa, form conduction t valence band, emitting light. In chapter 4.2 a wrinkled film is analyzed with amplitude $A$ and wavelength $\lambda$, consisting of a quantum well embedded between two barrier layers. The strain distribution of the wrinkled film is analyzed and the effect on the conduction and valence band profile is studied. Subsequently, the effect on the transition energies is analyzed for flat, concave and convex wrinkle profiles. Finally, the lateral carrier localization is studied and a comparison is made between the theoretical predictions and the experimentally-obtained photoluminescence measurements on wrinkled nanofilms that are partly bonded back to the substrate. The chapter closes with an outlook on the possible application areas of the strain engineering of the optical and electronic properties in thin film semiconductor film technology.

Mr. Cendula has demonstrated a profound understanding of the relation between film shape and morphology (rolling and wrinkling) and the internal strain distribution, allowing him to explore their effect on the electronic and optical properties, of large practical importance for micro-electronic and photonic systems. For these he used skills in a wide range of different scientific domains, including manufacturing, theoretical modeling and numerical simulation. Therefore, I recommend that the thesis of Mr. Cendula will be accepted, without any reservation.

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