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The stability of elastically strained nanorings and the formation of quantum dot molecules

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ABSTRACT

Self-assembled nanorings have recently been identified in a number of heteroepitaxially strained material systems. Under some circumstances these rings have been observed to break up into ring-shaped quantum dot molecules. A general non-linear model for the elastic strain energy of non-axisymmetric epitaxially strained nanostructures beyond the small slope assumption is developed. This model is then used to investigate the stability of strained nanorings evolving via surface diffusion subject to perturbations around their circumference. An expression for the fastest growing mode is determined and related to experimental observations. The model predicts a region of stability for rings below a critical radius, and also a region for larger rings which have a proportionally small thickness. The predictions of the model are shown to be consistent with the available results. For the heteroepitaxial InP on $In_{0.5}Ga_{0.5}P$ system investigated by Jevasuwan et al. (2013), the nanorings are found to be stable below a certain critical size. This is in good quantitative agreement with the model predictions. At larger sizes, the rings are unstable. The number of dots in the resulting quantum dot molecule is similar to the mode number for the fastest growing mode. Second order terms show that the number of dots is expected to reduce as the height of the ring increases in proportion to its thickness. The strained $In_{0.4}Ga_{0.6}As$ on GaAs nanorings of Hanke et al. (2007) are always stable and this is in accordance with the findings of the analysis. The Au nanorings of Ruffino et al. (2011) are stable as well, even as they expand during annealing. This observation is also shown to be consistent with the proposed model, which is expected to be useful in the design and tailoring of heteroepitaxial systems for the self-organisation of quantum dot molecules.

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1. Introduction

The unique nature of the electronic confinement in nanorings generates a range of quantum effects with possible exploitation in optical and magnetic applications and quantum computation devices (Ruffino et al., 2011; Somaschini et al., 2009).Their structure-dependent transmission/absorption spectra offer tunable plasmon resonances for optoelectronic devices (Sun et al., 2011; Fang et al., 2007; Kelf et al., 2011) and they have the potential for use in sensing devices (Huang et al., 2012; Aizpurua, 2003) and magnetic data storage devices (Yu et al., 2007). Nanorings have been produced by a variety of methods, such as the coating of nanoparticles (Larsson et al., 2007; He at al., 2010), the use of templates (Hobbs et al., 2004), the agglomeration of nanoclusters (Mishra et al., 2007), deposition on block co-polymers (Zahr and Blum, 2012; Wang et al., 2011), partial capping of quantum dots (Stoffel et al. 2009) and droplet epitaxy (Somaschini et al., 2009; Li and

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Yang, 2008). Most of these techniques are strain-free, except for the partial capping process, which typically involves significant intermixing producing a non-uniformly alloyed quantum ring (Hanke et al., 2007). Jevasuwan et al. (2011) have recently developed the technique of strained droplet epitaxy to produce nanorings of pure crystalline InP. In this process, liquid indium droplets are deposited onto an $In_{0.5}Ga_{0.5}P$ substrate, and subsequently exposed to a phosphorus beam. The In and P combine at the edge of the droplet to form crystalline InP. The InP continues to grow around the perimeter at the expense of the droplet, until the In droplet is entirely consumed leaving a InP nanoring. Above a certain size, the epitaxially strained nanoring becomes unstable (Jevasuwan et al., 2013) leading to the formation of a ring-shaped quantum dot molecule (QDM). This typically generates 6–14 quantum dots around the perimeter of the original droplet, depending on the geometry of the original nanoring. This does not occur in unstrained droplet epitaxy, for which the growth mechanisms are well understood (Li and Yang, 2008, 2009; Zhou et al., 2013). It is therefore reasonable to assume that the large epitaxial mismatch strain present in the InP on $In_{0.5}Ga_{0.5}P$ system is destabilising the nanoring. QDMs also have a variety of potential uses in optoelectronics (Wang et al., 2006) and quantum computation (Amlani et al., 1999; Li et al., 2003). Construction of these devices requires control over the size, shape, number and arrangement of the quantum dots within the QDM. A range of QDMs have been produced by other routes, mostly mediated by strain relief, due to preferential formation of quantum dots around mounds (Lee et al., 2008) or pits (Hu et al., 2012), producing bi-QDMs, quad-QDMs, hexa-QDMs and multi-QDMs.

This paper considers the stability of heteroepitaxial nanorings with the intention of determining the relationship between the geometry of a nanoring and its stability, with the expectation that the dominant unstable mode will determine the number of quantum dots in the resulting QDM. The main complexity of this analysis is determination of the elastic strain energy of the different perturbed nanoring structures. Section 2 considers the general case of a surface traction, expressed in polar co-ordinates, that is distributed over an isotropic elastic half-space. This analysis is expanded to second order terms in relation to the radial slope of the initial ring to account for large slope effects. In Section 3, a particular geometry for the nanoring is assumed, and an expression for the growth rate of different modal pertubations derived. The predictions of the model are presented in relation to experimental observations in Section 4.

2. Second order elastic strain energy of a generalised distribution of surface traction on an elastic half-space in polar coordinates

The following analysis is applicable to any axisymmetric or non-axisymmetric heteroepitaxial nanostructure whose height profile, $\xi(r, \phi) = h(r)g(\phi)$, is readily expressed in terms of separate functions of the polar coordinates, r and ϕ, where $g(\phi)$ is represented by a Fourier series. Here we assume that the initial symmetric shape, defined by the radial height profile $h(r)$, is subjected to a time-dependent asymmetric undulation in the hoop direction such that

$$\xi(\boldsymbol{r}, t) = \xi(r, \phi, t) = h(r)(1 + \eta(\mathrm{t}) \cos n\phi) \tag{1}$$

where n and $\eta(t)$ are the mode number and relative amplitude of the undulation. It is easy to show that the undulation conserves volume. The axisymmetric problem ($n = 0$) has been extensively studied by Shchukin et al. (2004) and Gill (2009). These linear analyses employed the small slope assumption (for slopes less than 0.15), whereby only first order surface tractions were considered. In this paper, second order terms are also included. This is partly because the observed slopes in the nanorings are relatively large (up to 0.3), but also because there are two interacting slopes in this case, the large radial slope of the initial nanoring, and the smaller slope in the hoop direction induced by the sinusoidal perturbation.

2.1. First and second order surface tractions

On a free surface, the normal, σ_N, and in-plane, σ_S and σ_T, surface tractions should be zero. In 3D polar co-ordinates (r, ϕ, z) the out-of-plane slopes are given by

$$\boldsymbol{s}(\boldsymbol{r}) = [s_r, s_\phi] = \left[\frac{\partial \xi}{\partial r}, \frac{1}{r}\frac{\partial \xi}{\partial \phi}\right] \tag{2}$$

where the components are in the radial and hoop directions respectively. The orientation of the surface defined by (1) is given by the corresponding radial, hoop and normal unit vectors

$$\boldsymbol{S} = \frac{[1, 0, s_r]}{\sqrt{1 + s_r^2}}, \quad \boldsymbol{T} = \frac{[0, 1, s_\phi]}{\sqrt{1 + s_\phi^2}}, \quad \boldsymbol{N} = \boldsymbol{S} \times \boldsymbol{T}. \tag{3}$$

To second order in the slopes these are

$$\boldsymbol{S} = \left[1 - \tfrac{1}{2}s_r^2, 0, s_r\right], \quad \boldsymbol{T} = \left[0, 1 - \tfrac{1}{2}s_\phi^2, s_\phi\right], \quad \boldsymbol{N} = \left[-s_r, -s_\phi, 1 - \tfrac{1}{2}(s_r^2 + s_\phi^2)\right]. \tag{4}$$

The surface tractions are

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